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Waste Management is designed for scientists, engineers, and managers, regardless of their discipline, who are involved scientific, technical and other issues related to solid waste management. Emphasis is placed on integrated approaches. The approaches require the blending of technical and non-technical factors. Although the dissemination and application of innovat technical information is extremely important, the implementation of sustainable waste management practices also require thorough understanding of the pertinent legal, social, economic, and regulatory issues involved.

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Editorial

First and foremost, I would like to take this opportunity to thank all of the referees who so selflessly review our manuscripts. As has now become a tradition at *Waste Management*, we are acknowledging all referees for the past year in a special section of this issue and would like to call to your attention a few referees who have been selected by the Editors as

Outstanding Referees for 2004:

Maria da Conceição M. Alvim Ferraz
Paul Cooper
Paul T. Imhoff
Steve Tamplin
Nickolas J. Themelis
Jamie VanGulck

On a different matter, during the last few weeks I have had the pleasure of participating in two very interesting and related conferences. One was held in Palma de Mallorca, Spain and the other one in Lake Toya, Hokkaido, Japan. Although the activities were held thousands of miles apart and in entirely different settings, the primary objectives of the meetings were uncannily similar. The meeting in Mallorca was sponsored by the Institute for Sustainable Resources and dealt primarily with "Sustainable Development and Islands – Resources and Waste Management". The second meeting was "The Third Intercontinental Landfill Research Symposium (ICLRS)" organized by distinguished colleagues from North Carolina State University, USA; Luleå University, Sweden; and Hokkaido University, Japan. The ICLRS has now become one of the premier events dealing with landfill research.

Interestingly, both meetings were held in well-known island resorts. In addition, both meetings dealt, in one way or another, with "sustainability" and with "resource management" within the context of solid waste management.

Those of us who have had the opportunity to work on projects related to solid waste management in islands understand the difficulty and importance of careful analyses prior to implementation of a waste management system. The management of solid wastes is a complex problem under most circumstances; the complexity is magnified several times in islands. This is particularly so in small, tourist settings which attract hundreds and sometimes thousands of visitors each month.

The conference in Toya Lake was made even more meaningful to us by holding our annual Editors meeting prior to the conference. The meeting was attended by the majority of the Editors, some members of the Editorial Advisory Group, as well as several members of the Editorial Board.

Participants at the Editors Meeting in Lake Toya, Japan

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The event in Palma was attended by many professionals from a variety of islands from throughout the world as well as by several key authorities from different countries. One of the illustrious participants was Mr. George Pullicino, Minister of Environment of Malta. Mr. Pullicino made an insigthful contribution to the Opening Session of the Congress which very aptly summarizes a number of issues facing islands and as such we would like to share his presentation with you.

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A view of the setting at Palma de Mallorca, Spain

Opening address

Distinguished guests, ladies and gentlemen.

One sought after resource in an insular community similar to Malta is space. The population of Malta is in the region of 397,000 inhabitants. Its population density is 1257 persons per square km, compared to the 170 persons per square km of the Balearic Islands. This makes space a commodity in continuous demand in Malta.

The subject that this conference addresses, that is Sustainable Development, timely coincides with the development of a "Sustainable Development Strategy for Malta". This Strategy is one crucial initiative that my Government has recently embarked upon and the process for formulating this plan is currently in the consultation phase. The Strategy is due for publication during the first quarter of 2005.

However, more importantly, this conference brings together two important issues for discussion that more often than not are competing at loggerheads with each other in communities with similar characteristics to Malta. These issues are space and the management of waste. The latter is an activity that unfortunately requires much usable land and void to be managed in an environmentally acceptable way.

In a country generating approximately 1.6 million tonnes of waste annually, space is quickly taken up for managing waste. Having recently joined the European Union, Malta is still synchronising its legal obligations as far as waste issues are concerned. Such obligations not only bestow extended responsibilities but also require fresh ground for the development of new facilities.

About 200,000 tonnes of the total waste generated is municipal waste with the remaining fraction being inert material from the construction industry. Most of the municipal waste is currently landfilled with some 4000 tonnes of second-class compost being recovered through the *Eco-pod* system. The biodegradable fraction of the municipal solid waste amounts to about 60%. On the other hand, all inert material generated in Malta is currently being used for the rehabilitation of disused quarries around Malta with most of the reclaimed land being used for agricultural purposes. Concurrent to this, the potential for a land reclamation project is being assessed. Through the assistance of foreign experts, a site selection exercise is being initiated, which will lead to the choice of a preferred site being thoroughly researched for this purpose.

Whilst waste management and space continue to compete and whilst Government tries to maintain an even balance between environment, social and economic implications as a result of this conflict, the generation of waste remains a clear indication of how much our community remains inefficient in its every day activity. This inefficiency is a clear daily reminder that resources continue to be wasted and remediation action is called for.

To partly address this issue, the Maltese Government has recently launched what is commonly referred to as an eco-contribution. This eco-contribution, imposed on a number of products/materials that are problematic when in the general waste stream, seeks to set a fiscal penalty on an item if its producer/importer puts it on the market with a complete disregard of collecting it

from the consumer once it reaches its end-of-life. If the product/material participates in an authorised 'take-back' scheme, then it becomes exempt from paying the contribution depending on how successful the scheme is in its recovery. This exemption is a sufficient incentive for industry to shoulder its responsibility towards better management of resources through the recovery of material from waste.

In an island state with little or no resources available to it, waste becomes a usable resource if managed satisfactorily. Sound waste management practices become a source for creating *green jobs*. There are impediments associated with treating waste on islands due to economies of scale. Such constraints may become more relevant when considering exporting waste. These, coupled by the fact that the sea isolates an island from available technologies on a continent, may make the export of waste a tougher feat to overcome. As such, Malta welcomes any initiative that seeks to bridge services as a means of connecting islands to overcome similar burdens to those referred to above.

Although safeguarding competition is a fundamental principle of the enlarged European Union, Malta strongly believes that this should not come at the expense of the environment. This is being said in view of the fact that Malta, as part of its accession agreement, has had to lift its ban on one-way beverage packaging. This ban was seen as a barrier to competition irrespective of the fact that the present recovery scheme for two-way packaging achieved some 95% recovery of all units placed on the market. As a result of this change in regulation, it is anticipated that during the transition period all two-way beverage packaging will switch to one-way packaging with significant environmental implications.

It is with pleasure to be able to participate in the opening of this congress especially since Maltese delegates, with a responsibility in waste management and under the responsibility of my Ministry, will be participating actively in the proceedings of this event. The similarities of other insular communities may thus be shared whilst diverse strategies adopted to overcome challenges may be compared.

I wish you all a fruitful and inspiring encounter.

George Pullicino
Minister of Environment, Malta

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Characterization and recovery of mercury from spent fluorescent lamps

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Abstract

Fluorescent lamps rely on mercury as the source of ultraviolet radiation for the production of visible light. Partitioning of mercury among vapor phase, loose phosphor powders produced during breaking and washing steps, glass matrices, phosphor powders attached on the glass and aluminum end caps was examined from simulated laboratory lamp recycling tests for different types of spent and new fluorescent lamps. Mercury concentrations in lamp glasses taken from commercial lamp recyclers were also analyzed for comparison with the simulated results of spent and new lamps of different types. The mercury content of the glass from spent lamps was highly variable depending on the lamp type and manufacturer; the median values of the mercury concentration in glasses for spent 26- (T8) and 38-mm (T12) diameter fluorescent lamps were approximately 30 and 45 μg/g, respectively. The average mercury concentration of samples taken from recycler A was 29.6 μg/g, which was about 64% of median value measured from the spent T12 lamps. Over 94% of total mercury in lamps remained either as a component of phosphor powders attached inside the lamp or in glass matrices. New T12 lamps had a higher partitioning percentage of elemental mercury in the vapor phase (0.17%) than spent T12 lamps (0.04%), while spent lamps had higher partitioning percentages of mercury resided on end-caps and phosphor powders detached from the breaking and washing steps. The TCLP values of simulated all lamp-glasses and samples obtained from recyclers were higher than the limit of LDR standard (0.025 mg/L). After investigating acid treatment and high temperature treatment as mercury reclamation techniques, it was found that heating provided the most effective mercury capture. Although the initial mercury concentrations of individual sample were different, the mercury concentrations after 1 h exposure at 100 °C were below 4 μg/g for all samples (i.e., <1% remaining). Therefore, it is recommended that heating be used for recovering mercury from spent fluorescent lamps.
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1. Introduction

The presence of mercury in the environment is a persistent and increasing problem. Since fluorescent lamps, which rely on mercury for their operation, are more energy-efficient than incandescent lamps (Hildenbrand et al., 2000; Thaler et al., 1995), their extensive use over the years has caused growing concerns over their proper disposal. The National Electrical Manufactures Association (NEMA) has projected that approximately 680 million fluorescent lamps will be disposed of in the United States in 2004 (NEMA, 2000). Although the total amount of mercury in fluorescent lamps varied significantly depending on the type of lamp and year of manufacture, the amount of mercury in fluorescent lamps has decreased over time due to the efforts to decrease environmental contamination. The mercury content of spent fluorescent lamps has been reported to be between 0.72 and 115 mg/lamp with an average mercury content of about 30 mg/lamp in 1994 (Battye et al., 1994; Truesdale et al., 1993) NEMA reported that the average mercury content of a 4-foot lamp was 11.6 mg in 1999.

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However, it has been found that many fluorescent lamps contain sufficient quantities of mercury to fail the toxicity characteristics for mercury when they are disposed. Due to persistent mismanagement of lamps, the Environmental Protection Agency (EPA) recently published the new hazardous waste lamp rule that adds waste lamps to the federal list of universal wastes, which are regulated under the Resource Conservation and Recovery Act (RCRA) (EPA, 1999). Furthermore, land disposal restriction (LDR) treatment requirements for characteristic wastes went into effect on May 8, 1992. Thus, before hazardous wastes containing mercury can be landfilled, they must be treated to below the LDR standards. The LDR standard specified in 40 CFR 268.40 for lamps that are mercury characteristic hazardous wastes is 0.025 mg/L, as determined by the TCLP test (Walter, 2001). One way to avoid the treatment of fluorescent lamps to LDR standards is to recycle the lamps.

Most lamp recyclers in the United States employ the dry recycling process which generates four products: mercury-contaminated phosphor powder, mercury-contaminated filters, crushed glass, and aluminum end caps. The entire system is operated under negative pressure to minimize mercury emissions to the atmosphere (Tanel et al., 1998). The spent fluorescent lamps are first broken in a crushing unit. During crushing, a vacuum system placed under crusher grates collects the air and crushed materials, preventing the mercury from escaping through the feed tube. The materials are passed through a cyclone separator, in which aluminum end caps and broken glass are separated and phosphor powder containing mercury is transferred to an enclosed auger conveyer. The elemental mercury released in the vapor phase is captured by carbon filters, which are ultimately sent out for mercury reclamation. Mercury-contaminated phosphor powder and filters containing elemental mercury are shipped as hazardous waste to a mercury refining company for retorting to recover elemental mercury and phosphor powder. These mercury-containing wastes are placed in a retort and heated above the boiling point of mercury (375 °C) for 4–20 h. The vaporized mercury is condensed in the scrubber and then collected in a decanter. Additional treatments such as nitric acid bubbling may be required to remove impurities from the mercury before it can be reused. The crushed glass is usually sent for reuse in other applications such as the manufacture of fiberglass insulation, road material or a multitude of other products. Aluminum end caps are sold to aluminum manufacturers to smelt into aluminum sheet or other products for resale as raw material.

Although the entire system of dry recycling procedure can minimize mercury emissions to the atmosphere, the effect of mercury adsorbed onto the glass on its recyclability and public health has not been investigated to date. There are also few studies on how much mercury is partitioned to the different compartments of spent and new fluorescent lamps.

Accordingly, in this study, wet chemical analysis (WCA) methods were utilized to achieve the following objectives: (1) determine partitioning of mercury among the vapor phase, loose phosphor produced during breaking and washing steps, lamp glass matrix, phosphor powder attached on the glass, and aluminum end caps for different types of spent and new fluorescent lamps; (2) compare mercury concentrations in glasses taken from lamp recyclers with the results of simulated laboratory recycling tests; (3) evaluate the effectiveness of mercury recovery methods from lamp components, such as acid solutions and heating process for extracting mercury from spent fluorescent lamps.

2. Materials and methods

Different types of spent fluorescent lamps were obtained from the University of Wisconsin-Madison Physical Plant. New lamps, manufactured by several companies, were purchased in retail stores in Madison, Wisconsin. Recycled glass samples were obtained from two commercial fluorescent lamp recyclers located in Wisconsin for TCLP tests and the amount of the total mercury in the glass samples was measured. The spent and new fluorescent lamps were used to determine partitioning of mercury in the following phases: (1) vaporized elemental mercury; (2) mercury released from phosphor powders, which were physically detached during lamp breaking and washing with deionized water; (3) mercury in the phosphor powder attached in the glass; (4) mercury adsorbed into the glass matrices; and (5) mercury adsorbed onto the end caps.

Specifications of spent and new fluorescent lamps tested in the study are summarized in Table 1. Two different types of lamps were tested: T8 and T12. T8 lamps are 26 mm in diameter and range from 600 to 1800 mm in length while T12 lamps are 38 mm in diameter and can range from 600 to 2400 mm in length. The T8 linear fluorescent lamp is the modern and energy efficient alternative to the T12 lamp. In this study, different types of spent T8, spent T12, and new T12 were selected with the same length of 1800 mm. Three different types of spent T8 were tested. New and spent T12 lamps manufactured by two companies were selected because their consumption is larger than other products in Wisconsin.

All mercury analyses were based on Method 7471B (Mercury in Solid or Semisolid Waste – Manual Cold Vapor Technique) from EPA's "Test Methods for Evaluating Solid Waste – Physical/Chemical Method," (SW-846) (EPA, 1998). In this study, it was not possible to obtain reproducible results from total mercury analysis because Method 7471B specifies a sample size of 0.6 g which is too small to represent the total sample. There-

Table 1
Specification of different types of spent and new fluorescent lamps

Types	Code	Specification	Mass (g)	Total Hg (mg/lamp)
Spent lamps				
T8	U-T8-P-H-1	F32T8/TL841 made in Holland	189 9	6.04
	U-T8-P-H-2	F32T8/TL841 made in Holland	189.3	4.44
	U-T8-P-H-3	F32T8/TL841 made in Holland	189.4	8.50
	U-T8-P-U-1	F32T8/TL741 made in USA	188.5	9.46
	U-T8-P-U-2	F32T8/TL741 made in USA	188.9	9.89
	U-T8-P-U-3	F32T8/TL741 made in USA	187.5	–
	U-T8-P-A-1	F32T8/TL841 alto collection	189.3	2.74
	U-T8-P-A-2	F32T8/TL841 alto collection	189.7	1.73
T12	U-T12-G-1	Shoplift F/40, UPC 43168-98103	279.3	10.29
	U-T12-G-2	Shoplift F/40, UPC 43168-98103	297.9	22.19
	U-T12-G-3	Shoplift F/40, UPC 43168-98103	301.4	7.99
	U-T12-G-4	Shoplift F/40, UPC 43168-98103	307.7	21.86
	U-T12-G-5	F40 RES 40 W, residential light	285.3	12.11
	U-T12-S-1	F40/CWX 40 W, cool white deluxe	284.0	16.48
	U-T12-S-2	F40/CWX 40 W, cool white deluxe	286.4	6.61
	U-T12-P-1	F40CW/RS/EW-II, 34 W, 046677-107413, USA	261.5	15.65
New lamps				
T12	N-T12-W-1	Realite full spectrum F40T12/FS, Made in Germany	301.4	25.35
	N-T12-W-2	Realite full spectrum F40T12/FS, Made in Germany	307.7	13.1
	N-T12-S-1	Designer warm white plus, F40/DWWP 40W USA	284.0	5.69
	N-T12-S-2	Designer warm white plus, F40/DWWP 40W USA	286.4	1.96
	N-T12-G-1	Residential light F40RES 40W	261.5	5.07
	N-T12-G-2	Residential light F40RES 40W	285.3	14.27
	N-T12-SE	Economy F25T12/CW 25W	300.0	2.41

fore, several modifications to Method 7471B were made for total mercury analysis so as to obtain reliable results for the entire weight of the lamp. The mercury analysis of each component of fluorescent lamps is described below.

2.1. Sample preparation and partitioning of mercury

Each lamp was weighed after being wiped clean with deionized water. After weighing, each lamp was placed onto a 140×25 cm laboratory bench paper. A dip scratch was made with a glass-cutter on the center of the fluorescent lamp. The elemental mercury in the vapor phase was captured by a collection system that was composed of a wide-mouth, cone shaped funnel (105 mm ID) connected to a peristaltic pump with a teflon tube. The teflon tube was immersed into 100-mL of mixed acid solution. The mixed acid solution was prepared with nitric acid 5% (v/v) and hydrochloric acid 5% (v/v) following the recommendation of Dominski (1985). Before crushing the lamp, the peristaltic pump was set on maximum speed. The lamp was broken into two parts with the stainless steel bar directly under the collection funnel. After collecting the mercury in the vapor phase into the mixed acid solution, the solution was analyzed for mercury immediately with the manual cold vapor method. The broken lamp was then separated into its component parts for testing. First of all, the following experiments were conducted to investigate how much mercury can be released from the loose phosphor powder produced through breaking the lamp and washing the inside of fluorescent lamps with deionized water. The inside of the lamp was washed with about 50 mL of deionized water for 30 min. The solution was then collected in a 100-mL volumetric flask. The mixed acid solution of hydrochloric acid and nitric acid was added into this solution to be 5% (v/v) for each acid solution. Next, the total volume was adjusted to 100 mL with additional deionized water. The solution was stirred at room temperature for 24 h before analysis by the manual cold vapor method. The lamp glasses were dried under vacuum at room temperature for 4 h without collection of mercury and wrapped with laboratory bench paper and then shattered into 2–3 in. pieces with a hammer. The glass pieces were inserted into the grinder and the lid was closed tightly. The glass was gently pulverized into small particles for 10 min. in order to obtain more homogeneous samples for mercury analyses. The pulverized particles were collected in a pre-cleaned 300-mL capped vessel. Before analysis, the samples were preserved in a refrigerator at 4 °C. The pulverized glass samples were used for several mercury analysis including TCLP and mercury extraction tests.

The separated aluminum end caps free of phosphor powder from each lamp were weighed and analyzed for mercury using method 7471B. Both end caps were added to a pre-cleaned 300-mL capped-vessel and digested with a mixture of 25 mL of reagent water, 25

mL of aqua regia, and 12.5 mL of potassium permanganate solution, which can be designated as the digestion mixture. Aqua regia was prepared immediately before use by carefully adding three volumes of concentrated HCl to one volume of concentrated HNO_3. Potassium permanganate was prepared at 5% based on the weight to volume basis. The capped vessel was mixed using a shaker for 18 ± 2 h at room temperature. The supernatant was then filtered with a 0.45-μm pore size filter and diluted with mixed acid solution of hydrochloric acid (5%, v/v) and nitric acid (5%, v/v) to stay within the range of the standard curve.

To analyze the total mercury concentration of the lamp glasses, approximately 50 g of the pulverized glass from each lamp tested in this study and lamp glasses obtained from the recyclers was weighed and placed in a 300-mL capped vessel. The mercury extraction and analysis methods were the same as those of end-caps described above.

Since total mercury concentration of lamp glasses includes both mercury immobilized into phosphor powder attached on the glass, and mercury partitioned to the glass matrices, the following experiments were performed to determine how much mercury was partitioned to the glass matrices. Only spent T12 fluorescent lamps were used for this experiment. About 20 g of the pulverized glass was placed into a 500-mL volumetric flask. Then, 400 mL of deionized water was poured over the glass sample. The flask was mixed vigorously using a shaker for 18 ± 2 h at room temperature. The supernatant including the phosphor powder was discarded, and this procedure was repeated several times to remove all phosphor powder until the glass samples became transparent. Since mercury concentrations in the entire lamp and washed glass were measured, the mercury concentration in the supernatent was not measured. In order to measure the mercury concentrations of glasses without phosphor powders, these samples were dried at room temperature for 24 h. Raposo et al. (2003) showed that mercury extinction started above 250 °C in the TD (thermal desorption) profile of mercury within a glass sample of a spent fluorescent lamp. This might be caused by a strong linkage of mercury into the matrix of glass. Therefore, the mercury could not be released during the drying step at room temperature for 24 h. Samples were weighed again and placed in a 300-mL capped vessel. As described previously, these samples were digested with the digestion mixture. The mercury extraction procedure and analysis were the same as the previous experimental procedure of mercury analysis for glass.

2.2. Toxicity characteristics leaching procedure

In order to perform a modified TCLP test, approximately 20 g of pulverized glass from each lamp and glass sample obtained from recycler was placed in a 500-mL volumetric flask. Next, 400 mL of the acetate buffer solution with a pH of 4.93 ± 0.05 was poured into the volumetric flask in order to meet the ratio of 20 (mass of glass (g)/volume of acetate buffer solution (mL)). The flask was shaken in a shaker for 18 ± 2 h at room temperature. After shaking, the supernatant was filtered with a 0.60-μm pore size glass micro-fiber filter and diluted with mixed acid solution of hydrochloric acid (5%, v/v) and nitric acid (5%, v/v) to be in the range of the standard curve. The diluted solution was analyzed with the manual cold vapor method of atomic absorption spectrophotometer (AAS).

2.3. Evaluation of mercury extraction tests

Mercury extraction tests were performed to evaluate the efficiency of two methods of extracting mercury from lamp components: acid extraction and thermal desorption. For evaluation of acid extraction, one of the spent T8 lamps was used for the mercury extraction tests. The mass of the lamp was 187.5 g. The mercury concentration resided onto the lamp glass was 62.42 μg/g in the previous analysis. Approximately 10 g of pulverized glass was placed in a pre-cleaned 300-mL capped vessel. Next, 100 mL of different concentrations and types of acid solutions were poured into the capped vessel. The acid solutions for this experiment were nitric acid and the mixture of nitric and hydrochloric acid solution (1:1 volume ratio). The concentrations of these two different acid solutions were 0.5, 1, 3, 5, 8, and 12% (v/v), including the control. The capped vessel was mixed in a shaker for 18 ± 2 h at room temperature. The supernatant was filtered with a 0.45-μm pore size filter and diluted with the mixed acid solution of hydrochloric acid (5%) and nitric acid (5%). The diluted solutions were analyzed with the manual cold vapor method of AAS.

The efficiency of mercury extraction by heating was measured at various temperatures. Spent T8 lamps and a sample taken from a recycler were used for this experiment. Approximately 150 g of pulverized glass was evenly distributed through an area of 15 × 15 cm on aluminum foil. The glass samples were placed into a furnace (Isotherm Programmable Muffle Furnace, Fisher Scientific®). After 1 h at a specific temperature, 20 g was taken from the original sample. The temperatures tested were 100, 200, 300, 400 and 500 °C.

2.4. Mercury analysis

All standard solutions were prepared by diluting the mercury stock solution of 1000 mg/L. The standard curve was fitted with the 2nd degree polynomial method, resulting in a very reliable R^2 value of over 0.999. The measurement time and pre-read delay time were 8 and

90 s, respectively. The vaporized mercury was quantified at the wavelength of 253.7 nm. As a quality control check on the analysis, blank and EPA reference standards were analyzed with each set of samples.

3. Experimental results

Box-and-whisker plots were used to present the results of mercury analyses. It provides a snapshot of a data set using five statistics: the minimum (10th percentile), the 25th percentile, the median, the 75th percentile and the maximum (90th percentile). A line inside the box shows the position of the median. The plot also has lines extending from the box showing the approximate regions occupied by outliers and extreme values. The location of the median line and the relative length of the whiskers indicate how symmetrical the data are. If the median lies far from the center of the box or one whisker is much longer than the other, the data distribution is considered skewed.

3.1. Mercury in vapor phase

The box-and-whisker plot of the elemental mercury concentrations in the vapor phase is shown in Fig. 1(a). The median value of vapor phase mercury for the new T12 lamps (4.0 μg) was greater than that of the spent T12 lamps (2.9 μg). These values were much smaller than other reported values: 0.04 mg (NEMA, 2000) and 0.018 mg (Aucott et al., 2003). This result might be caused by different experimental setup and measurement methods for the release of elemental mercury. The mercury in the vapor phase from the spent T8 lamps was not detected. Thus, from the above experimental results, it can be surmised that elemental mercury in vapor phase could be consumed and partitioned to other compartments such as end caps or glass matrices during the service periods.

3.2. Mercury in end caps

The box-and-whisker plot of mercury concentration in the end caps of fluorescent lamps is shown in Fig. 1(b). The median value for the spent T8 lamps was about 8 μg/g, showing the greatest value among the various types of lamps. Although the spent T12 lamps had the median value of about 1 μg/g in the end caps, their data distribution was skewed to higher values (~13 μg/g at 90th percentile), suggesting a high variance. Meanwhile, the mercury concentration in end caps for the new T12 lamps was lowest (a median value of 0.1 μg/g) among other types of lamps tested. For the case of spent T8 lamps, although the mercury in the vapor phase was not detected, a large amount of mercury was detected in end caps (Fig. 1(b)). On the other hand,

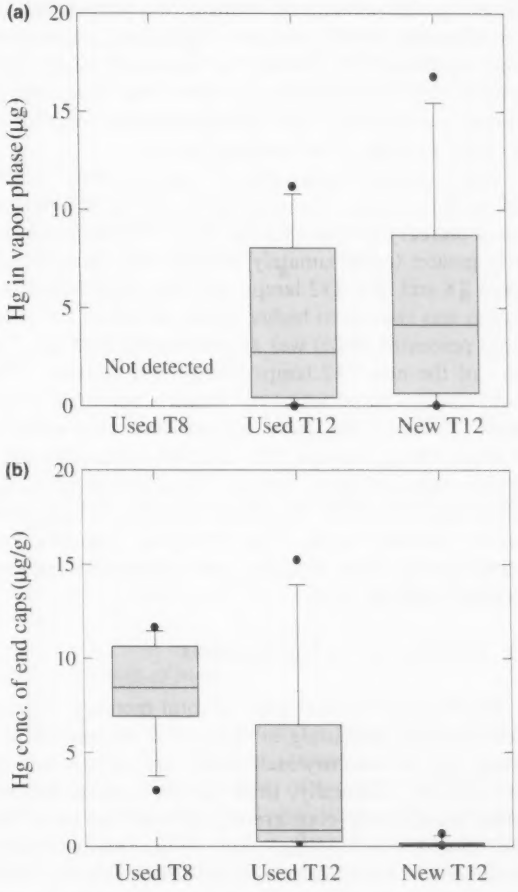

Fig. 1. (a) Box-and-whisker plot of mercury in the vapor phase. (b) Box-and-whisker plot of mercury concentrations of end caps.

the new T12 lamps had a reverse situation. Thus, the elemental mercury from the vapor phase appeared to be gradually adsorbed onto aluminum end caps or matrices of lamp glass over time during service. This result can be explained by the following mechanisms. First, the elemental mercury excessively added during manufacturing is partitioned into phosphor powder and it became unavailable during the operation of the lamp due to the oxidative reactions with phosphor powder (Hildenbrand et al., 2000). Second, elemental mercury can also be consumed by penetrating the oxide layer such as soda-lime glass or aluminum end-caps, and eventually residing into the matrix of metal oxide (Hildenbrand et al., 2000; Hildenbrand et al., 2003).

3.3. Mercury analysis of the loose phosphor powders produced from breaking and washing steps

Since the mercury-containing phosphor powders can be mobile through air and liquid phases during lamp

breaking, their detachability might be an important factor of public health concerns. Therefore, experiments were conducted to measure the masses of mercury extracted from the phosphor powders that were released during the breaking step and subsequently leached out through washing with deionized water.

The box-and-whisker plot of mercury mass in 100-mL of the washing solution is shown in Fig. 2. The median of mercury amounts for the spent T12 lamps was not only greater (approximately 500 μg) than those for the spent T8 and new T12 lamps, but also their data distribution was skewed to higher values, in which the maximum percentile (90%) was approximately 2000 μg. The data of the new T12 lamps had a high variance, even if its median value was very small. Compared to the spent T12 lamps, the spent T8 lamps had a low variance of about 400 μg between 10th and 90th percentiles and a median value of about 100 μg. Thus, compared to the other lamps, the spent T12 lamps appeared to have more mobile mercury in the loose phosphor powders produced during lamp breaking and subsequent leaching through washing steps.

3.4. Total mercury in broken glasses

The box-and-whisker plot of total mercury concentration (μg of mercury/g of lamp) and amounts in the lamps (mg of mercury/each lamp) are shown in Fig. 3(a) and (b). Generally, total mercury concentrations varied significantly even among different lamps of the same model, which corresponded to the results of other studies (EPA, 1992; Iskander et al., 1994; NEMA, 1994). Although the median of mercury concentrations (30 μg/g) in spent T8 lamps was lower than that of spent T12 lamps, the interquantile ranges (IQRs) between the 10th and 90th percentiles for both the spent T8 and T12 lamps were almost the same (~40 μg/g), indicating large variances for both types of lamps. Meanwhile, the new T12 lamps had a lower median value (~20 μg/g) than the spent T8 and T12 lamps. However, the data distribution (IQRs of 10–90th percentiles) of new T12 lamps was larger than spent T8 and T12 lamps.

The median value of the mercury mass per lamp for the spent T8 lamps (~6 mg/lamp) was calculated to be about 60% lower than that of the spent T12 lamps (~15 mg/lamp) because the spent T8 lamps usually weighed less than spent T12 lamps (Fig. 3(b)). In addition, the IQR between the 10 and 90th percentiles of spent T8 lamps (8 mg/lamp) was lower than that of spent T12 lamps (16 mg/lamp), indicating that the spent T12 lamps had a higher variance of mercury amounts based on each lamp. Among the spent T8 lamps, U-T8-P-A lamps (Alto® lamps) had the lowest mercury masses of below 3 mg/lamp.

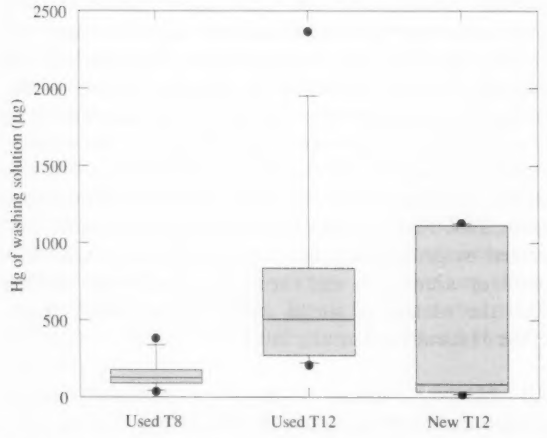

Fig. 2. Box-and-whisker plot of mercury in wash solution including the loose phosphor powders obtained during lamp breaking and washing steps.

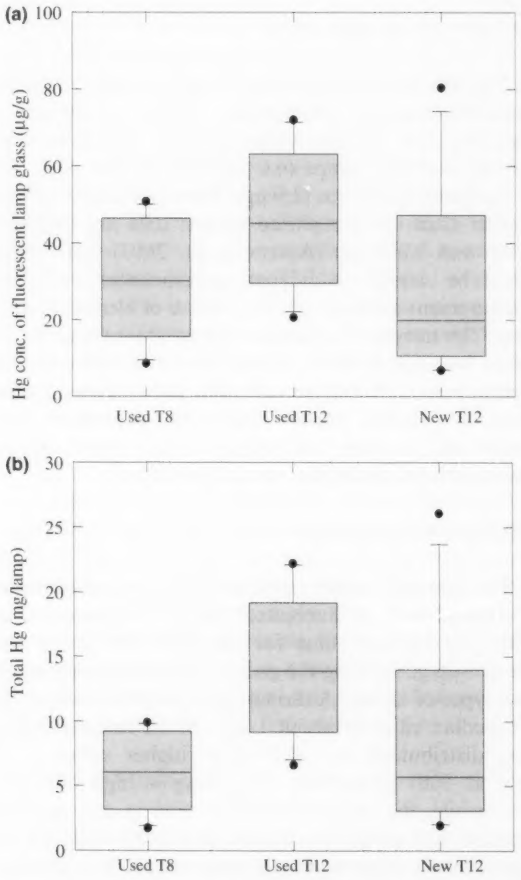

Fig. 3. (a) Box-and-whisker plot of total mercury concentration (μg of mercury/g of lamp). (b) total mercury amounts in the lamps (mg of mercury/each lamp).

The average and standard deviation of mercury concentrations in glass samples taken from recyclers A and B that employ dry recycling processes were 29.6 ± 0.071 and 3.8 ± 0.279 μg/g, respectively, indicating significant differences among recyclers. In addition, the phosphor powders still remained on the recycler glass although the amounts of phosphor powders were not measured. Therefore, it can be suggested that mercury exists on the glasses and residual phosphor powder at various levels depending on the separation processes used by recyclers, and that the separation process of lamp recyclers cannot remove the mercury completely.

Assuming that all crushed glass produced from dry separation system are sent for reuse in manufacturing using heating processes and that all mercury is released to the air, the expected amounts of released-mercury per year in the United States (0.15–1.2 tons/year) could be calculated from the obtained results of mercury concentrations. For this calculation, the following figures were used: projected lamps-disposed number (680 millions) and 25% of recycled option at 2004 (NEMA, 2000), as well as average mass of lamp (238.5 g) for T8 and T12 types. The expected maximum value of 1.2 tons/year is about 10% of the total mercury contained in lamps (13 tons) forecasted for 2004 (NEMA, 2000).

3.5. Estimation of mercury adsorbed into the glass matrix of spent lamps

In order to estimate mercury concentrations in glass matrices, pure glass samples were digested with the digestion mixture after all phosphor powders attached onto the glass were completely removed by the procedure previously described. Since the ionized and excited mercury atoms are reactive, they can react with all components of lamps during the operating time, leading to irreversible fixation (Hildenbrand et al., 2003). In general, mercury ions can diffuse into the glass matrix in the presence of an electric field because Na ions in soda-lime glass are mobile to support the flow of Hg ions (Hildenbrand et al., 2000). However, the mercury partitioning in the glass matrix can be obstructed by coating with phosphor; Doughty et al. (1995) and Thaler et al. (1995) have empirically observed that the mercury concentration on the glass was approximately proportional to the square root of time and inversely proportional to the quantity of phosphor. Fig. 4 shows the results of mercury concentrations on several types of spent T12 lamps with and without phosphor powder. Although the mercury concentrations in lamps varied significantly, the concentrations of mercury adsorbed into glass matrices were very close (~3.5 μg/g) for all samples. These data show that most of the mercury is present in the phosphor powder residue on the glass, and that the lamp glass adsorbs about the same amount of mercury regardless of the mercury in the phosphor powder.

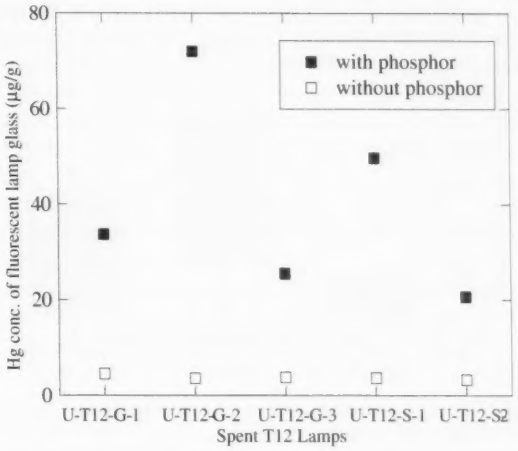

Fig. 4. Concentrations of mercury absorbed into the glass matrices (without phosphor) and mercury on the glass (with phosphor) of spent T12 lamps.

3.6. Partitioning of mercury

Table 2 shows the partitioning percentages of mercury mass to four different components for different types of lamps. Median values of mercury mass for each component were used for computing partitioning percentages. From Table 2, it is clear that most mercury in all different types of lamps was partitioned to glass matrices and the phosphor powders attached onto glasses. New T12 lamps had higher partitioning percentage of elemental mercury in the vapor phase (0.17%) than spent T12 lamps (0.04%). However, the mercury was not detected in the vapor phase of spent T8 lamps. Spent T8 lamps had the highest partitioning percentage of mercury concentration in end caps (2.07%) than that of spent T12 (0.50%) and new T12 (0.09%). Spent T12 lamps showed approximately twice-greater partitioning percentage of mercury mass in wash solution (5.34%) than those of spent T8 and new T12 lamps.

Table 2
Partitioning percentages of mercury mass for different components of fluorescent lamps

Components	Spent T8 lamps (%)	Spent T12 lamps (%)	New T12 lamps (%)
End caps	2.07	0.50	0.09
Loose phosphor powders obtained during breaking and washing steps	2.86	5.34	2.72
Vapor phase	N/D[a]	0.04	0.17
Phosphor powders remained on the glass and glass matrices	95.08	94.12	97.02

[a] N/D, not detected.

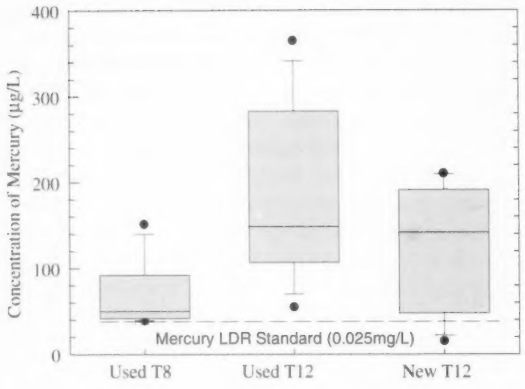

Fig. 5. Box-and-whisker plot of TCLP tests (the dashed line is the limit of TC and LDRs for mercury, 200 μg/L).

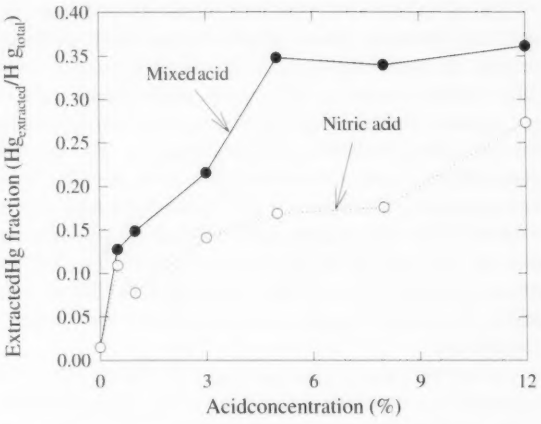

Fig. 6. Mercury extraction with nitric acid and mixed acid (hydrochloric acid + nitric acid).

3.7. TCLP tests

The box-and-whisker plot of modified TCLP results for all different types of lamps is shown in Fig. 5. Most data except the 10th percentile of TCLP for new T12 lamps were higher than the limits of the LDR standard for mercury of 0.025 mg/L (Walter, 2001). The TCLP data of the spent T12 lamps varied significantly from about 50–360 μg/L. Although the median values of TCLP tests for both the spent and new T12 lamps were approximately 150 μg/L, the data set of the spent T12 lamps was skewed toward higher values, in which a 75th percentile is higher than 200 μg/L. For the new T12 lamps, the 90th percentile and maximum values of TCLP results were over 200 μg/L, while their data were skewed to the lower range. The IQRs of the TCLP results for both the spent and new T12 type lamps were approximately 150 μg/L of differences between 25th and 75th percentiles, indicating that the TCLP results were highly variable. Since the mercury form in lamps at the time of manufacture is predominantly metallic which has a very low solubility in the TCLP test and the metallic mercury is converted to more soluble complexes through conversion into other compounds in the lamp (Nesting et al., 2001), TCLP results for spent T12 would be expected to be higher than new T12. However, in this study, this cannot be definitively explained by the oxidative change of elemental mercury during the operation, because the median value of total mercury for new T12 lamps (mg of Hg/lamp) was lower than that of spent T12 lamps. Although the TCLP data of spent T8 lamps had a skewed distribution toward higher values, they had a lower median value of 50 μg/L and IQR of about 50 μg/L of differences between the 25th and 75th percentiles, showing much more consistent and reliable data.

For the glass residues from the fluorescent lamp recyclers, the average and standard deviation values of TCLP results were 129.87 ± 1.99 μg/L (recycler A) and 104.47 ± 8.85 μg/L (recycler B), respectively. Although these values are below the median values of spent and new T12 lamps simulated in this study, they are still higher than the limit of LDR standard. Accordingly, lamp glasses should be managed as a decharacterized waste and sent off-site to a Treatment Storage and Disposal Facility (TSDF) for further treatment (Walter, 2001).

3.8. Mercury extraction tests with different types of acid solution

The mercury fraction extracted by different types and concentrations of acid solution is shown in Fig. 6. For both acid solutions, the extracted mercury fraction seemed to increase with increase in acid concentrations. The mixture of nitric and hydrochloric acid solution had higher mercury extraction from the glass than the nitric acid only solution. The extracted mercury fraction increased sharply to about 35% up to 5% of the mixed acid solution. At >5% of mixed acid concentration, however, mercury extraction was not improved. The maximum extracted mercury fractions were 36% for mixed acid and 28% for nitric acid, respectively, indicating that acid extraction is not effective for mercury recovery from fluorescent lamps.

3.9. Effect of temperature on mercury release from glass

The remaining mercury concentrations in the glass after 1 h of exposure at different temperatures are shown in Fig. 7. Although the initial mercury concentrations of individual samples were different, the mercury concentrations after 1 h of exposure at 100 °C were below 4 μg/g for all samples (i.e., <1% remaining). The remaining mercury concentrations gradually decreased with

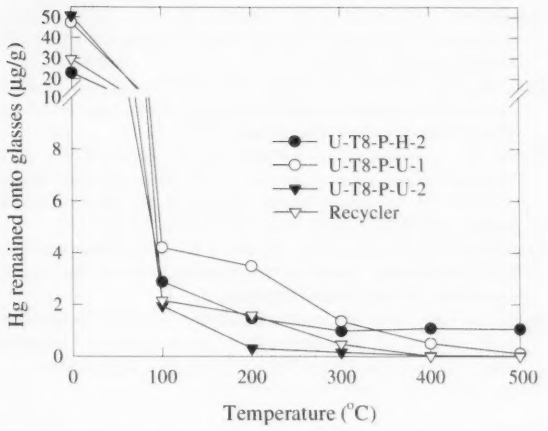

Fig. 7. Temperature effect for mercury extraction from fluorescent lamp glasses.

the increase in temperature. Glass taken from the recyclers decreased sharply from about 30 μg/g of mercury to 2 μg/g after 1 h of exposure at 100 °C. After heating up to 400 °C, mercury was not detected. However, the U-T8-P-H-2 did not have a complete removal of mercury even after treating at 500 °C, indicating that mercury can be strongly bound to glass matrices.

Considering two mercury extraction techniques, heating is simple, reliable, and efficient for recovering mercury from spent fluorescent lamp glasses. It is recommended that all fluorescent lamp recyclers use heating not acid extraction for mercury recovery from lamps.

4. Conclusions

A series of mercury analyses from various parts of T8 and T12 fluorescent lamps was performed to determine the partitioning of mercury in five different components of new and spent fluorescent lamps – vapor phase, loose phosphor produced during breaking and washing steps, end caps, and the glass matrices. Glass samples were also obtained from two lamp recycling companies and compared with glasses of tested lamps. The following conclusions were drawn:

1. Through oxidative reactions with phosphor powder and penetration mechanisms, elemental mercury in vapor phase has been partitioned to other compartments such as end caps or glass matrices during the service although the partitioning was different depending on the lamp types.
2. Since the mercury-containing phosphor powders can be mobile through air and liquid phases when lamps are broken, the detachability of mercury-containing phosphor powders might be an important factor of public health concerns. From detachability tests of mercury-containing phosphor powders, the mercury in phosphor powders of spent T12 lamps appeared to be more mobile than the spent T8 lamps.
3. Total mercury concentrations and the amounts of mercury varied significantly even among different lamps of the same model, which was in agreement with the results of other studies. Among the spent T8 lamps, Alto® lamps had the lowest mercury masses of below 3 mg/lamp.
4. Mercury exists in the phosphor powder residue on the glasses at various levels, depending on the separation processes used by recyclers. The separation processes employed by most lamp recyclers cannot remove the phosphor powder and mercury on lamp glasses completely. Accordingly, when lamp glasses are recycled, the mercury residue on glasses will volatilize and may be emitted to the atmosphere.
5. Compared with the acid washing, the heating process was efficient for recovering mercury partitioned on the glass. The mercury concentrations after 1 h of exposure at 100 °C decreased to below approximately 4 μg/g. The remaining mercury was gradually volatilized with an increase in temperature. Above 400 °C, mercury was recovered almost completely, although some types of glass can strongly complex with mercury.

Acknowledgements

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Influence of distance on the motivation and frequency of household recycling

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Abstract

People choose to participate in recycling for a variety of reasons. This study analyzes the relationship between the frequency of selective separation and general refuse disposal, and the influence on the recycling habit of the walking distance to drop off the materials. The methodology employed was one of personal interviews in the street, the study population being a region in the north of Spain (Principality of Asturias). More than one thousand people participated in this survey carried out in 2002. Five hypotheses related to three variables (the frequency for depositing recycling materials and general refuse, the distance to recycling and general bins, and the recycling habit) were tested using different statistical tests. Results show that the people who frequently go to the bins to dispose of general refuse are more likely to recycle some product at home, and in most cases, as the distance to the recycling bins decreases, the number of fractions that citizens separate and collect at home increases. Most of the results obtained have been compared with other previous in literature.
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1. Introduction

It is becoming more and more common practice for products that have been used and thrown away by users at the end of their useful life to be recovered. Consumer collaboration, however, is crucial to achieve this goal. It is the consumer who is responsible for carrying out primary separation at home, distinguishing between containers (paper/cardboard, glass, and light packaging – such as cans and plastic containers), separating these from the rest of the refuse and dropping them off in the corresponding bin.

Several collection systems exist for recyclables. Excluding pay-as-you-throw systems due to their limited implementation in Europe, the main systems are (Lesceu, 2000):

1. *Curbside collection*. In this case, the bins are set out by residents next to those for general refuse. This is the most costly system, since it means a greater number of collection points and is therefore a service that requires more time and higher resources; although collection occurs very near the source of the waste. It is very convenient for users and is aimed at assuring a high participation on their part.
2. *Neighborhood collection*. This method involves defining an area where a set of large bins are located for the collection of different items. These are usually placed in public squares or other spacious areas, with easy access for collection trucks; still, they are located further from the source, reducing the convenience to residents.
3. *Clean point*. Also called a green point, this is a site designed for the selective drop-off of products that are not collected by means of the previous systems (hazardous residues, home appliances, computer equipment, etc.), namely, a center for the selective

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Table 1
Recycling motivation results from similar studies

Reference	Aim	Geographic setting	Methodology	Main results
Belton et al. (1994)	To ascertain why citizens decide whether to separate household waste or not	Glasgow (Scotland)	Personal questionnaire in the street	67% of citizens separate glass, 55% paper, 29% plastic and 19% aluminum cans. Reasons are: 15% economic, 2% social and 25% ecological.
Callan and Thomas (1997)	To estimate the effectiveness of practices that encourage recycling	Massachusetts (USA)	Case studies for counties	Impact of socio-economic factors in recycling
PCAESG (1999)	To ascertain recycling tendencies and education	UK	Questionnaire	Paper (57%) and glass (50.7%) are the most frequently separated materials versus 23% for light containers. Distance and ease of access to drop-offs are the main reasons for recycling motivation.
Jenkins et al. (2000)	To assess the environmental impact of two different recycling programs for household waste	USA	e-mail questionnaire	74.6% recycle newspapers, 66.5% glass bottles, 63.2% aluminum and 54.2% plastic bottles. The main difficulties are transportation and storage of separated household waste at home.
Gonnerman et al. (2000)	To show citizens' acceptance of recycling programs	Iowa (USA)	Phone questionnaire	51.7% return empty containers to the shop or deposit center due to money return. 83% are aware of some recycling program.
Speirs and Tucker (2001)	To ascertain the profile of citizens that make special trips for recycling	Glasgow and Ayrshire (Scotland)	Personal questionnaire in supermarket	22% of citizens make special trips only for recycling. They tend to shorten the distance and make drop-offs less frequently.
Owens et al. (2002)	To obtain information on recycling behavior for selected populations	Athens-Clarke (Georgia, USA)	Questionnaire	45% of citizens are involved in recycling. Annual household income, home-ownership status and level or education were associated with recycling efficiency.
Domina and Koch (2002)	To ascertain the convenience of recycling frequency and variables that could be used as indicators in the prediction of recycling behavior.	USA	Mail questionnaire	Access, shopping behavior, family size and income are significant predictors of recycling activity.

reception and storage of waste. These are even further from the source of waste and are the least convenient to users.

2. Prior studies

Many regional studies have been conducted over the last decade into the topic of recycling motivation in different countries. Worth highlighting among these is the one directed by the *Packaging Consumer Awareness and Educations Steering Group* (PCAESG, 1999), developed in Great Britain, which concludes that paper and glass are the recyclable materials that consumers reprocess most (with recycling rates of 57% and 50.5%, respectively, as opposed to 23% for light packaging). At the same time, the study found that 60% of consumers separate at most one distinct item in their homes. Finally, they found that proximity and ease of access to the sites for depositing separated waste stand out among the motives that stimulate recycling.

Another study was carried out in Glasgow that evaluated the use that people made of the bins for selectively separating refuse (Belton et al., 1994). This study describes when the different types of selective separation bins for the different wastes began to be used and the reasons why people have this habit or not. A detailed questionnaire was developed for actual users of the bins and another for people chosen at random near to shopping areas, whether they were users or not. In all, 251 questionnaires were completed, from which it was seen that 67% separated glass, 55% paper, 29% plastic and 19% aluminum cans. The average distance to the bins was 0.54 km for those who went on foot (39% chose this option) and between 0.68 and 2.64 km if they went by car (59% of those interviewed). The main motives for recycling were economic (in 15% of cases), social (2%) and ecological (25%). Non-recyclers were found to be young people of low social class. Among the reasons they gave were distance (22%) and that they had not thought about recycling before (12%). In response to a question on when separated wastes were deposited, 55% did so on their way to the shops, 23% went out especially to do this, and 3% did so on their way to work.

Researching this topic also in Scotland, Speirs and Tucker (2001) analyzed the profile of the people who make special trips just to recycle. According to their results, 22% of recyclers make special trips, mainly short trips (45% of them drop off paper and glass together). They look for *Clean Points* with parking space and easy access.

In the USA, Gonnerman et al. (2000) carried out another study along the same lines in the State of Iowa. They randomly contacted a total of 830 adults over the age of 18 by telephone. A large percentage of these admitted to returning empty drinks bottles to the store or shop where they habitually bought them or to a deposit center, although 51.7% admitted to doing so only to get back the value of the deposit. This study, prepared by the *Iowa Department of Natural Resources*, also reports that 85% of the people surveyed were aware of one or other of the recycling programs carried out in their community.

With respect to the factors that encourage selective collection, social influences and altruistic and regulatory factors are some of the reasons why certain communities develop strong recycling habits (Vining and Ebrero, 1992; Ewing, 2001). The major drawbacks, on the other hand, are the effort needed (Oskamp et al., 1991; McCarty and Shrum, 1994), the space needed (Vining and Ebrero, 1990; Vining et al., 1992; Boldero, 1995) and cost (Ewing, 2001). Other reasons have to do with established habits or insufficient information (Pieters, 1991). Table 1 presents a comparison of some of the results from the literature including other that have not been commented in detail before.

3. Geographical scope and the aims of the present research

The Principality of Asturias, the area under study, is located in Northern Spain (see Fig. 1). It is a mountainous

Fig. 1. Location of the principality of Asturias in Northern Spain.

Table 2
Schema of the hypotheses tested

	Distance for depositing general refuse	Frequency of depositing general refuse	Number of fractions recycled
Frequency of depositing recycled materials	–	H1	H2
Distance to recycling bins	H3	–	H5
Recycling habit (Y/N)	H4a	H4b	–

Table 3
Questionnaire employed in the study

Data related to the collection of general refuse

1. How frequently do you drop off refuse in the bin?

   everyday every two days twice a week once a week less than once a week

2. What types of refuse do you separate and how frequently?

	Paper	Glass	Light packaging	Batteries	Other_____
every day					
every two days					
twice a week					
once a week					
twice per month					
less than twice per month					

3. Time it takes from your home to the nearest bin:

   - For refuse: less than 1 minute between 1 and 5 minutes between 5 and 10 minutes
     more than 10 minutes do not know
   - For recycled items : less than 1 minute between 1 and 5 minutes
     between 5 and 10 minutes more than 10 minutes do not know

Data related to the recycling mentality

4. For those refuse items that you do separate (or would separate), mark the importance you give to the following reasons for separating using the following scale.

Totally Disagree | 1 | 2 | 3 | 4 | 5 | 6 | 7 | Totally Agree

Neither agree nor disagree

	1	2	3	4	5	6	7
Environmental awareness							
Duty as a citizen							
Proximity of bins to user's place of residence							
Distance between general refuse bins and selective ones							
Complying with regulations							
Other.....							

5. For those items that you do not separate (or would not separate), mark the reasons using the same scale.

	1	2	3	4	5	6	7
Distance between general refuse bins and selective ones							
Lack of space in user's home							
Time needed to carry waste to bins							
Finding the effort not useful							
Other							

region with a population of 1,087,885 citizens and a density of 101 people per square kilometre, usually divided in 3 areas (Western – 113,596 inhabitants, mainly farmers –, Central – the most populated area, where the government and industrial facilities are located, with 919,042 – and Eastern – with 55,247, mainly tourist villages).

COGERSA, a public company owned by the municipalities of the Central area of the region, is responsible

for the management of solid waste. This company carries out all the processing of solid waste (collection, treatment and disposal) in the region. Two different systems are used for collection. In the first one, waste is collected (mainly through *curbside bins*) and transported for its disposal at COGERSA's landfill. In the second one, when the county is too far from the landfill, pretreatment plants (i.e., transfer stations) are used in which the waste is compacted in order to reduce its volume before being sent to the landfill. A total of 1,600 tons of municipal solid waste (MSW) is disposed of daily (COGERSA, 2003).

COGERSA also has a collection system for recycling that has been functioning since 1994 and which is composed of *neighborhood collection* systems and *clean points* and two classification plants (for processing paper and packaging, respectively). In 2002, 18,803 tons of paper, 6389 tons of glass and 3347 tons of light packaging were collected by this company (COGERSA, 2003). The Asturian waste management system is the same as those found in other Spanish regions.

3.1. Hypotheses tested

The distance required to drop off the refuse has been mentioned on different occasions as an important influence on recycling behaviour. The aim of this research study is to analyze the behavior of the consumers of packaged products with respect to the frequency and distance to travel to selectively separate household waste. The study focused on the Principality of Asturias, analyzing the profile of consumers and their concern for the environment. The research tested the following five null hypotheses (see Table 2):

- *Hypothesis 1 (H1):* The frequency with which a person drops off refuse in the general refuse bin, and the periodicity with which he or she carries out the same action with the separated fraction, are independent.
- *Hypothesis 2 (H2):* The frequency with which people take different items to the bins is similar in all three cases (paper, glass and light packaging).
- *Hypothesis 3 (H3):* The time needed by a person who separates at least one item to take items to the recycling bins is independent of the time he or she needs to drop off general refuse.
- *Hypothesis 4 (H4):* (a) The fact that the bins (including those for general refuse) are to be found near to the place of residence, and (b) the frequency with which general refuse is usually dropped off in the bin, are independent of the habit of separating waste at home or not.
- *Hypothesis 5 (H5):* The number of different items recycled is independent of the distance that has to be covered to drop off the refuse in the bin.

4. Methodology

A personal survey was chosen to carry out this research. Segmentation of the sample within the Principality of Asturias was carried out by means of two successive divisions. Initially, the sample (targeted to be some 1000 interviews) was distributed among the three areas of Asturias in proportion to the population, the total number of surveys to be carried out in each of these areas being, respectively, 111, 901 and 54 (1066 interviews in total). Subsequently, the surveys were distributed amongst the different municipalities or counties in each of the three areas, taking into account the number of bins installed in each of these. In theory, and assuming no bias in the interviewees who took the survey, with a sample of this size, we may state at a level of 95 percent certainty that the results have a statistical precision of plus or minus 3 percentage points with respect to what they would be if the entire adult population had been polled with complete accuracy.

The survey was carried out in the street in the morning and afternoon of weekdays by the authors of this paper. The interviewees were randomly chosen as they passed by in the street subsequent to their prior consent. The pedestrians rarely refused to answer because the survey only required a couple of minutes. The questionnaire employed in the interview is shown in Table 3. The statistical methodology used in each case is commented on in the next section.

5. Results and analysis

From Fig. 2 we can see that paper is dropped off once a week or more frequently in 59.8% of cases, while the proportion for glass is 49.4%, 36.0% for light containers and 14.9% for batteries. These results for glass and paper are similar to those in the sample used by Speirs and Tucker (2001), where 45% of the analyzed recyclers made their trip at least once a week. Similar results are reported by Ball and Lawson (1990) in a study that took place in Scotland in which 52% of recyclers go to the drop-off points once or twice a month, showing similarities between our sample and those of other populations.

It has already been mentioned that the frequency with which both general and separated refuse is dropped off is one of the variables of interest in this research study. It might be that the more often general refuse is taken to the bin (as was seen in the review of the literature, the individual might try and make the most of the trip), the more often waste will be dropped off in the selective collection bins.

Regarding this factor, the first of the goals of this research (H1) was to analyze the frequency with which general refuse and recycled items are dropped off in the bins (questions 1 and 2, Table 3). Three testes were

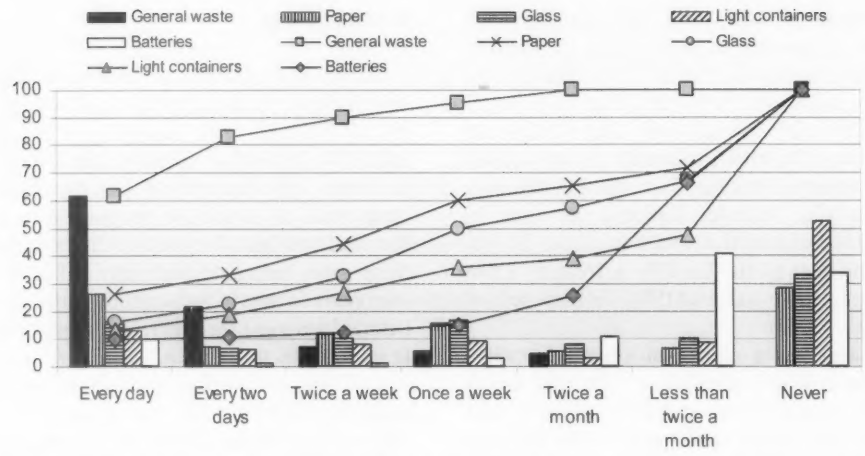

Fig. 2. Frequency (in percentage) with which people drop off the different recycled items in the bins (bars: absolute value; line, cumulative).

carried out using contingency tables, Pearson's χ^2 test, in which the relationship of independence between both variables (frequencies) is rejected (Table 4). However, it is not possible to prove the existence of any type of statistically significant linear correlation (see Pearson correlation, last column, Table 4) between the frequency of carrying non-separated household waste to the bin and the periodicity with which recycled items (paper, glass and light packaging) are dropped off in the selective collection bins. From the sample, general refuse is dropped off in the bins every day, while recycled items are dropped off less frequently. All these data are shown in Fig. 2.

The aim of the second hypothesis (H2) was to check whether all the recycled items are dropped off in the selective collection bins with similar frequencies (question 2, Table 3). To do so, a t test was carried out for related samples, the detailed results of which are presented in Table 5, in which we can observe if the frequencies of depositing the different pairs of recycled items are correlated. As shown by the t statistic in the second column of Table 5, the hypothesis of equality of means must be rejected, paper being the most frequently deposited fraction (mean rate of once a week), while light containers are least frequently deposited (mean rate of twice a month), probably due to the more common use of other types of containers. The mean rate for depositing of glass is 1.5 weeks. Ewing (2001) claims that paper is generally deposited more frequently because it presents fewer storage problems until drop-off and collection than light packaging.

In order to test the third hypothesis (H3), only those cases of people who selectively separate waste were chosen (question 3, Table 3). A new contingency table was analyzed to check the existence of independence between the time needed to drop off recycled items in the bin and the time spent in carrying out said action with general refuse. The majority of people have both general refuse and selective bins at a distance of less than 5 minutes from their homes, though in the case of selective bins they normally take more than 1 minute. The results indicate the rejection of H3, and show that there is a linear

Table 4
Contingency tables (total percentages) of the frequency of depositing general refuse and recycled items (significance level in brackets)

	General refuse	
	χ^2	Pearson's correlation
Paper	83.688 (0.000)	−0.006 (0.851)
Glass	64.992 (0.000)	0.046 (0.134)
Light packaging	77.729 (0.000)	0.057 (0.063)

Scale: 1: every day; 2: every two days; 3: twice a week; 4: once a week; 5: twice a month; 6: less than twice a month; 7: never (for general refuse – columns-, 6 = 7 = never).

Table 5
Student t test for the frequencies of depositing the different recycled items of household waste and the average frequency of depositing the three items

	Correlation	t
Paper–glass	0.641*	−4.031*
Paper-packaging	0.541*	8.566*
Glass-packaging	0.571*	13.097*

* Significance level of 0.00.

Table 6
Contingency table (total percentages) of the time spent dropping off refuse in the bins and the habit of separating waste (significance level in brackets)

Separate	Time needed to carry waste to bins				χ^2
	<1′	1′–5′	5′–10′	>10′	
Yes	44.0	33.6	3.8	0.7	6.552 (0.088)
No	8.0	9.1	0.7	0.2	

Table 7
Contingency table (total percentages) of the frequency of carrying refuse to the bins, and the habit of separating waste (significance level in brackets)

Separate	Frequency of carrying refuse to bins (%)						χ^2
	Every day	Every two days	Twice a week	Once a week	Twice a month	Less than twice a month	
Yes	51.1	16.5	5.3	5.0	4.0	0.1	12.331 (0.031)
No	10.4	5.1	1.4	0.6	0.4	0.1	

Table 8
People's motives for selectively separating and not separating waste at home (a 7-point Liker scale was employed: 1-absolute disagree; 4-indifference; 7-absolute agree)

Separating	Reasons	Mean value	Standard deviation	Confidence interval (95%)
Yes	Environment awareness	6.05	1.3	(5.96; 6.13)
	Duty as a citizen	5.26	1.6	(5.16; 5.37)
	Proximity of containers to users place of residence	4.90	1.83	(4.78; 5.03)
	Distance amongst general waste containers and selective ones	4.56	1.96	(4.43; 4.69)
	Complying regulations	3.43	1.93	(3.30; 3.56)
No	Distance amongst general containers and selective ones	4.78	2.12	(4.61; 4.94)
	Lacking space in users home	4.67	2.13	(4.51; 4.83)
	Tim for carrying waste to containers	3.66	1.94	(3.51; 3.80)
	Finding the effort not useful	3.08	1.85	(2.94; 3.22)

Table 9
Contingency table (total percentages) of the time spent dropping off refuse in the bins and the number of items recycled (significance level in brackets)

Number of items recycled	Time needed to carry waste to bins				χ^2
	<1′	1′–5′	5′–10′	>10′	
0	7.8	9.0	0.6	0.2	24.819 (0.016)
1	5.9	3.2	0.7	0.1	
2	7.4	5.5	0.9	0.1	
3	**9.0**	**9.4**	1.3	0.3	
4	**21.9**	**15.6**	1.0	0.2	

relation between the time needed to take waste to both general refuse and selective bins ($\chi^2 = 317.718$, and Pearson correlation = 0.413, both with a significance level of 0.00).

We consider this to be a consequence of the environmental laws passed in the country and the efforts by municipalities to promote recycling, which has meant the deployment of thousands of recycling bins. Other authors have likewise analyzed the distance issue. For example, Ball and Lawson (1990) reported that more time is employed in other countries to carry refuse to the general or selective bins is higher, mainly since people have the habit of traveling for another motive, such as shopping, work or entertainment, to drop off recycling. When making use of the trip for other purposes and dropping off the refuse on the way, the citizen is willing to go further, and in fact does so, than if the displacement were solely to deposit refuse in the bin.

For the next hypotheses, the proximity of bins to the household was measured by means of the time taken by each person to take their refuse to the bin (questions 3 and 2, Table 3). Using contingency tables, we conclude that in this case it is not possible to reject, at a confidence level of 95%, the independence between the recycling habit and the time needed to drop off this material (value 0.088 > 0.5 in Table 6).

On the other hand, once more using a contingency table to compare the frequency of disposing of general refuse and the recycling habit (using the answers to questions 1 and 2 in Table 3), we can reject the independence between both factors at a confidence level of 95% (Table 7). A correlation analysis carried out did not show a significant linear relation between the frequency of depositing refuse in the bin and the habit of separating waste material at home.

Note should be taken here that according to our results (Question 4 in the interview sheet), the people with "a recycling habit" consider their main stimulus to be their concern about their surrounding environment, considering this task to be the duty of every citizen. The succeeding motives (Table 8) for selectively separating waste at home are in fact the proximity of the corresponding bins to the household, followed by the proximity of these to the bins where general refuse is dropped off. In contrast, the predominant reasons for not separating at home or not separating certain items

are the distance of selective collection bins with respect to the bins where the rest of the refuse is dropped off, and the space needed at home for selective separation.

This last finding may explain why, when analysing H4, it was seen that the people who often go to the general refuse bins are especially motivated to separate waste material at home. The problem of the lack of space at home for storing waste material is minimized when people have the habit of frequently disposing of this waste in the bins.

The fifth hypothesis (H5) analyzes the independence of the number of items recycled with respect to the time spent in dropping off the waste in the bin (questions 2 and 3 in Table 3). On the basis of the results obtained (Table 9), the hypothesis of independence between the number of recycled items separated at home and the time needed to go to the bins can be rejected (see χ^2 column). It can be seen in this table that the majority of people who separate three or four items spend less than five minutes in going from their home to the point where the bins are located; i.e., the efforts (measured as the number of fractions separated at home) of the people with recycling habit, are encouraged by the proximity of the bins to their homes.

6. Conclusions

This study contributes data relative to consumer attitudes with respect to selective separation at home. A personal survey was carried out on a sample of a thousand people with the aim of defining whether the population is aware of this topic and what the most significant factors are.

The results indicate that dependence exists between the type of separated waste and the frequency with which it is dropped off in the selective collection bins. Paper is the most frequently separated item, on average it is carried out at least once every week to the selective bin.

Our results support the common assumption that when citizens who are environmentally concerned have bins near to home, they appear to be willing to recycle more fractions than when they have to walk for a longer time to drop off the waste, due to the inconvenience that carrying the large volumes that this type of waste usually occupies supposes.

As we have seen in this sample, the importance of ease of access to the bins is obviously an incentive to recycle. Once more, this is also consistent with the results obtained by Speirs and Tucker (2001) in Scotland, the PCAESG (1999) in UK, and Domina and Koch (2002), confirming their conclusions for the Spanish case as well. However, in the Spanish case, given the relative proximity of the bins (generally at a walking distance), distance does not act as an encouraging/inhibiting factor for the recycling habit, but rather affects the number of fractions that are separated.

In the Spanish case (as a result of the laws passed by the Government in 1998 that obliged municipalities to increase the number of curbside bins), an environmentally concerned person normally does not expect to spend more than five minutes going with his or her bags to the selective collection bins. Because of the proximity of bins in the majority of cases, the time spent carrying items to selective collection bins is not appreciated by citizens as an important point for deciding between separating household waste or not. However, the distance to the selective recycling bins does affect the number of fractions that citizens separate at home. On the other hand, as in other studies, one of the main reasons given by people who did not have this habit is the large amount of storage space required at home to carry out this task.

We considered the system chosen to gather data in this study (street interviews) to be a very appropriate procedure for a theme in which we are all involved agents. It allows the opinion of a large number of citizens to be tested, while guaranteeing randomness. Moreover, given the short time needed to answer the questionnaire and face-to-face presence with the interviewer, participation is practically one hundred percent.

The results obtained with respect to the tendency to selectively separate waste at home in this region (similar to a certain extent to those obtained in other countries) points out to the different public administrations how an effort in bringing refuse bins closer to users acts as an effective incentive with respect to the number of materials recycled in the home. A cost/benefit analysis of the repercussions that this effort in assigning resources supposes for governments might be a question to evaluate in subsequent research studies in this field.

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Heavy metal content in soil reclaimed from a municipal solid waste landfill

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Abstract

Residues reclaimed from a municipal solid waste (MSW) landfill were characterized for the concentrations of a number of heavy metals. The residue fractions analyzed included a fine fraction (<0.425 mm), an intermediate fraction (>0.425 and <6.3 mm) and a fraction consisting of paper products that could ultimately degrade to a smaller size. The intermediate fraction appeared to be organic in nature, while the fine fraction was more soil-like. In general, the metal concentrations were greatest in the intermediate fraction and lowest in the fine fraction. The effect of sample age on the elemental content was also investigated. The concentrations of several elements were greater in older samples (sample ≈8 years in age) when compared to newer samples (sample ≈3 years in age). Limitations associated with the land application of residual soil (composed of the fine and intermediate fractions) were assessed by comparing measured concentrations to regulatory threshold values. In general, most metal concentrations were below regulatory thresholds for use in unrestricted settings. At the concentrations measured, however, several elements might limit reuse options, depending on which regulatory threshold serves as a benchmark. Elevated concentrations of arsenic presented the greatest limitation with respect to common US thresholds while elevated cadmium concentrations presented the greatest limitation when compared to UK thresholds. The source of the arsenic was determined to be the waste, not the cover soil.
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1. Introduction

Municipal solid waste (MSW) landfill reclamation is the mining of landfills to recover metal, glass, plastic and other combustibles, soil, and the landfill volume itself (Morelli, 1990). Approximately 55% of the MSW generated in the United States is landfilled annually (US EPA, 2002). Although the number of landfills is decreasing, the capacity has remained relatively constant because newer landfills are much larger than those built previously (US EPA, 2002). With the inception of bioreactor operation to promote rapid stabilization of the landfilled waste, the MSW landfill has been promoted as a potential biochemical waste treatment unit rather than a mere waste storage facility (Morelli, 1990; Spencer, 1990; Magnuson, 1991; Reinhart and Townsend, 1997). While landfill reclamation was originally conceived as a method to address groundwater contamination problems at old, unlined landfills, it has been proposed as a potentially integral part of future bioreactor landfills (Nelson, 1994; Reinhart and Townsend, 1997). Reclaiming stabilized waste offers potential economic benefits for landfill operators through the creation of additional disposal capacity.

The major end product from the treatment of waste in a bioreactor landfill, in addition to landfill gas, is the stabilized residual material. The stabilized residual materials consist of soil, medium-sized residue and oversized residue. Major components of the oversized residue fraction include plastic, metals, wood, leather and rubber. The cover soil and degraded organic waste have been reported to constitute ≈75% of the recovered

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material (US EPA, 1997a). Potential reuse options for the recovered soil include use as daily and intermediate landfill cover material and as construction fill (US EPA, 1997a). A number of other end uses may be possible, dictated by available markets, the quality of the material, and the regulatory framework for reuse. The quality issue that would most likely limit its use would be the presence of trace amounts of hazardous chemicals (Reinhart and Townsend, 1997). It is likely that most of the organic chemicals will eventually be destroyed in the biochemical environment of the landfill (Field et al., 1995; Reinhart and Townsend, 1997; Atuanya et al., 2000; Cohen and Speitel, 2001). Contaminants such as heavy metals, however, will remain in the waste unless leached out. Several investigations indicate that the migration of heavy metals is very low during the first decades after deposition compared to the accumulated amount (Finnveden, 1996). Several attenuating mechanisms that limit the mobility of metals in the landfill have been shown to exist (Gould et al., 1990). More than 90% of non-metals and more than 99.9% of metals are reported as still found in the residual solids at the beginning of the humic phase (Belevi and Baccini, 1989; Bozkurt et al., 1999).

Before stabilized materials reclaimed from a landfill are land applied, they should be characterized for heavy metals of environmental concern. Stabilized residues from a MSW test cell in Sweden were characterized for heavy metals (Flyhammar et al., 1998). Hg levels in stabilized residues from a MSW landfill in US have been reported (Earle et al., 1999). Residues passing through 19.1- and 9.5-mm screens from an aerobic bioreactor test cell in Georgia, US, were characterized for metals (Das et al., 2002). The results of these studies are limited because only a small number of samples were used to characterize the reclaimed material, which is very heterogeneous in nature. Moreover, the studies focused on a limited number of elements. There is need for a more comprehensive characterization of heavy metals in residual material recovered from MSW landfills in the United States. In addition to the opportunities presented as a result of bioreactor operations, the difficulty in siting new landfills will push municipalities to explore the possibility of mining old, unlined landfills to create new disposal capacity. The disposition of the reclaimed soils will have an impact of the feasibility of such efforts.

The primary objective of this study was to characterize reclaimed residues from a MSW landfill in Florida in terms of heavy metals. The human health risk associated with land application of reclaimed residual soil was assessed by comparing results to typical "risk-based" standards or guidelines for heavy metals in soils developed in the US and Europe (US EPA, 1996; MHSPE, 2000; DEFRA and Environment Agency, 2002). A secondary objective was to investigate the impact of the age of waste on metal concentrations in reclaimed residues.

2. Materials and methods

2.1. Site description

The Alachua County Southwest Landfill (ACSWL) is located in north central Florida. The site consists of a number of landfill units, including an 11-ha (27-acre) composite-lined unit that began receiving waste in 1988. Approximately 270 metric tons per day of waste were deposited in the lined unit between 1988 and 1998. A sandy soil was used as daily and intermediate cover at the site. A detailed description of the landfill unit can be found elsewhere (Townsend et al., 1995). In 1990, an infiltration pond leachate recirculation system was constructed and operated for 28 months in a section of the lined landfill to examine the effects of leachate addition on landfill stabilization (Townsend et al., 1996). In 1993, leachate was recirculated using 11 horizontal injection lines in a section adjacent to the leachate recirculation pond to evaluate the performance of horizontal injection lines for leachate recirculation (Townsend and Miller, 1998). Hg levels in stabilized residuals from part of this landfill unit and an adjacent unlined section have been reported (Earle et al., 1999).

Gas extraction wells were constructed with 20-cm (8-in.) perforated PVC pipe and installed after the landfill closed. The wells were drilled with a 75-cm (30-in.) bucket auger. During the process of drilling holes for gas wells, samples were collected at 1.5 m (5 ft) intervals to a maximum depth of 18 m (60 ft). Sample sizes ranged from 10 to 20 kg. A total of 128 solid waste samples were collected from nineteen gas wells. Eighty-nine samples from fourteen wells were processed and analyzed for metals. Each sample was double-bagged with large plastic trash bags, excess air was squeezed from the bags, and both bags were individually tied. All samples were transported to the laboratory where they were frozen until analysis. Sample ages were estimated from landfill records of waste placement.

2.2. Sample processing

Because of their heterogeneous nature, waste samples required processing prior to analyses. Fig. 1 schematically illustrates the sample-processing scheme. The moisture content of each sample was measured by drying the entire sample at 105 °C for 24–48 h, depending on the time needed for the sample to reach a constant weight. After drying, some of the samples were combined to reduce sample-processing time. Samples from consecutive depth locations of a gas well were combined to create composite samples for intervals of 3.3–5 m (10–15 ft). Samples were combined only if moisture contents were within ±10% and the samples were similar in appearance (an indicator of sample age). The process of combining samples resulted in 78 samples. Samples were then size-

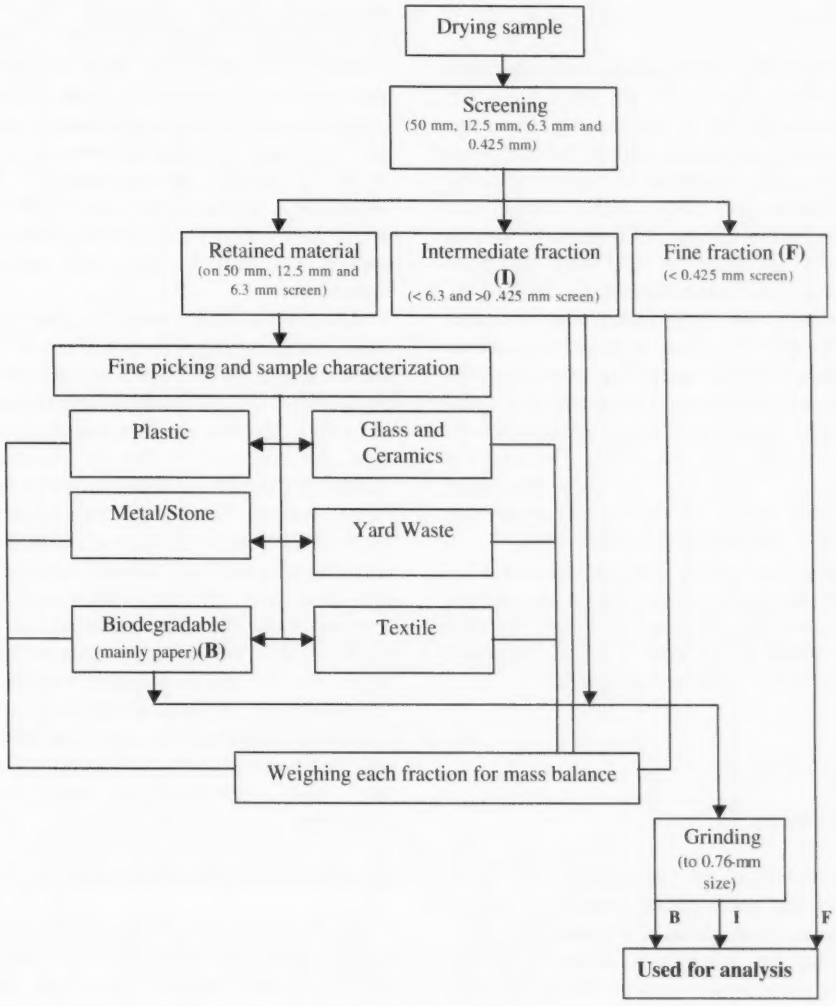

Fig. 1. Sample processing scheme.

separated using a shaker table with sieves of sizes 50, 12.5, 6.3 and 0.425 mm. The 50, 12.5 and 6.3 mm sieve sizes were selected based on reported typical screen sizes used for landfill reclamation (Reinhart and Townsend, 1997). The 0.425-mm screen was selected based on the results of study conducted by Miller et al. (1996), who reported that a 0.42-mm sieve allowed most of the cover soil to pass through (≈99%) while retaining a majority of the biodegradable material.

Sample fractions retained on the 50, 12.5 and 6.3 mm sieves were manually separated into the following six different categories: (i) paper; (ii) plastic; (iii) yard waste; (iv) glass and ceramics; (v) textiles; (vi) stone/metal. Only 25% of the sample (by weight) retained on the 12.5- and 6.3-mm sieves was used for the component categorization exercise. Each component was weighed. Representative samples were collected for the fraction that passed through the 0.425-mm sieve, referred to herein as the fine fraction; the fraction retained on the 0.425-mm sieve, referred to as the retained or intermediate fraction; and the fraction consisting of paper and paper products greater than 6.3 mm size, referred herein as the paper fraction. The paper fraction (in addition to the fine and retained fractions) was characterized for metals as this might ultimately degrade and become part of the screened residue of a reclamation activity. The intermediate and paper fractions were ground using a 2200-W Pulverisette, manufactured by Fritsch (Germany). All three fractions of each sample were refrigerated. The sample processing scheme used in this study was very similar to one reported by Miller et al. (1996) that was also used by Earle et al. (1999) for investigating Hg levels in MSW samples collected from the same site.

2.3. Sample analyses

All three fractions (fine, intermediate and paper fraction) were analyzed for Ag, Al, As, Ba, Be, Cd, Co, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Se, V and Zn. Total recoverable concentrations for all metals except mercury were measured using hot plate digestion with nitric acid following method 3050B (US EPA, 1995). Arsenic was analyzed using a Perkin–Elmer 5100 graphite furnace atomic absorption spectrometer (GFAA), equipped with Zeeman correction, following method 7060A (US EPA, 1995). Samples with high metal concentrations were diluted to fit within the linear region of the calibration curve. All other metals except Hg were analyzed using inductively coupled plasma atomic emission spectrometer (ICP-AES), Model 36 Thermo Elemental Inc., following method 6010B (US EPA, 1995). The total Hg concentrations in the three fractions were measured using the cold-vapor atomic absorption technique, following method 7471 of SW-846 (US EPA, 1995).

Quality assurance and quality control procedures included calibration checks, duplicates, blanks and sample spikes. Approximately 5% of the samples were digested in duplicate and spiked in duplicate to assess the precision and accuracy of the analytical procedures.

3. Results and discussion

3.1. Sample composition

The moisture content of all 128 samples was measured. After combining some of the samples to reduce the sample processing time, a total of seventy-eight samples were processed for component characterization. Fig. 2 presents the mean percentages of various constituents (by weight) in the 78 samples. The fine and intermediate fractions constituted ≈60% of the recovered material by weight (dry basis). The moisture content of the samples ranged from 9.3% to 51%, with a mean of 28% (wet weight basis). While no significant difference was observed between the mean moisture contents of samples collected from sections with and without leachate recirculation, samples from areas previously subjected to leachate recirculation did differ in physical appearance. Samples from areas where leachate recirculation had been practiced were much darker in color, smaller in particle size and more degraded in appearance.

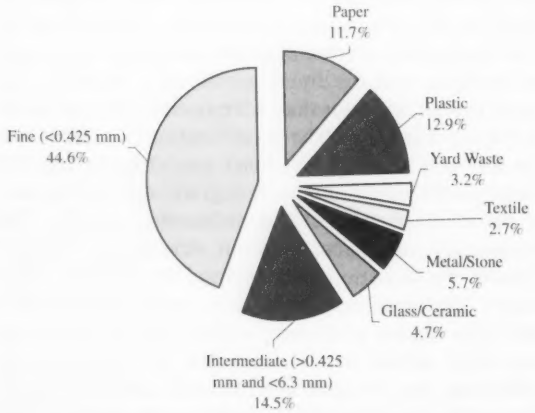

Fig. 2. Average composition of reclaimed material (78 samples).

The percentage (by weight) of the fine fraction in the MSW samples ranged from 6.6% to 94.5% with an arithmetic mean of 44.5%, which is comparable to 50% as reported for the waste samples from the same site (Earle et al., 1999). The fine fraction was composed primarily of sand, the cover soil at this site. The percentage (by dry weight) of the intermediate fraction in the MSW samples ranged from 2.7% to 73.2% with an arithmetic mean of 14.5%. The main constituent of the intermediate fraction was degraded organic material contaminated with broken glass and ceramic. The range of this fraction spanned wider than that reported by a previous study (Earle et al., 1999). The percentage (by weight) of the paper in the MSW samples ranged from 0.4% to 42.3% with the arithmetic mean of 11.7%. In addition to normal heterogeneity associated with MSW, factors such as decomposition and location of cover soil layers may also contribute to high sample-to-sample variability.

3.2. Elemental distribution in different fractions

In this section, metal concentrations in the fine, intermediate and paper fractions are presented and discussed. A few samples were lost during digestion/analysis process, so the concentration of all metals could not be obtained for every sample. The matrix spike recoveries for a majority of the metals were within 80–120%. Table 1 presents the method detection limit, number of samples analyzed, median, geometric mean, geometric standard deviation, and range of concentrations for each of the fractions (paper, intermediate and the fine). The coefficient of variation test indicated that for all the metals, the concentrations were not normally distributed (US EPA, 2000), and thus the median and the geometric mean were used to describe the central tendency of the data.

If the concentration of a metal was below the detection limit in less than 15% of the samples of a fraction (fine, intermediate, paper), then one-half of the detection limit was used to represent the concentration of that sample for statistical calculations and analyses. If the number of non-detects was between 15% and 50% of samples of a fraction, the mean and standard deviation were estimated using Cohen's method, an approach

Table 1
Metal concentrations in three fractions of stabilized MSW (mg/kg dry basis)

	Detection limit	Fine				Intermediate				Paper			
		Number of samples[a]	Median	GM ± GSD[b]	Range	Number of samples[a]	Median	GM ± GSD[b]	Range	Number of samples[a]	Median	GM ± GSD[b]	Range
Al[c]	0.004	77(77)	3.5	4.5 ± 0.0	1.9–76.1	78(78)	10.3	12 ± 0.0	2.5–169	78(78)	7.9	7.8 ± 0.0	1.7–39.3
Ag	1.5	77(19)	1.5[e]	–	<1.5–23.8	78(20)	1.5[e]	–	<1.5–24.6	78(16)	1.5[e]	–	<1.5–37.3
As	0.2	77(77)	1.5	1.4 ± 2.3	0.2–10.1	76(76)	4.2	4.5 ± 2.1	1.1–58.6	74(74)	4.3	5.2 ± 3.1	0.5–188
Ba	0.4	77(77)	16.1	18.3 ± 1.9	8.2–70	78(78)	50.8	54.1 ± 1.9	13.8–681	78(78)	37.3	38.6 ± 1.7	16.1–360
Be	0.1	77(17)	0.1[e]	–	<0.1–0.3	78(37)	0.1[e]	–	<0.1–0.8	78(15)	0.1[e]	–	<0.1–0.4
Cd	0.3	77(55)	0.6	0.5 ± 3.7[d]	<0.3–13.8	78(72)	1.8	1.9 ± 2.7	<0.3–40	78(74)	1.0	1.2 ± 2.7	0.3–90
Co	0.5	77(43)	0.7	0.6 ± 3.1[d]	<0.5–32.9	78(77)	2.6	2.9 ± 2.2	<0.5–86.7	78(74)	1.7	1.9 ± 2.2	<0.5–45.2
Cr	0.9	77(77)	8.6	9.5 ± 2.2	2.5–151	78(78)	39.4	44.0 ± 2.1	9.5–531	78(78)	23.9	24.7 ± 1.9	5.8–187
Cu	0.7	77(76)	16.9	16.9 ± 3.2	0.7–170	78(78)	89.7	105 ± 3.1	5.8–5530	78(78)	47.2	52.9 ± 2.0	12.5–352
Fe[c]	0.002	77(77)	3.0	3.2 ± 0.0	0.8–28.2	78(78)	14.8	14.6 ± 0.0	4.6–61.8	78(78)	5.1	5.6 ± 0.0	1–51.5
Hg	0.04	75(75)	0.1	0.13 ± 2.87	<0.04–1.8	74(74)	0.2	0.24 ± 3.33	0.04–9.0	71(71)	0.1	0.16 ± 2.66	0.04–1.7
Mn	0.6	77(77)	47.8	44.2 ± 3.0	6.2–993	78(78)	181	209 ± 2.6	50.4–14700	78(78)	78.0	93.5 ± 1.8	35.7–457
Ni	0.8	77(77)	5.0	5.0 ± 2.8	0.8–340	78(78)	18.8	21.2 ± 2.2	6.4–743	78(78)	10.7	11.7 ± 2.4	2.1–491
Pb	2	77(68)	9.0	10.8 ± 3.4	<2.0–297	78(71)	25.5	26.6 ± 4.8	<2.0–3250	78(56)	9.3	5.9 ± 7.9[d]	<2.0–627
Se	2.5	77(49)	4.2	3.7 ± 2.5[d]	<2.5–16.1	78(46)	3.0	3.1 ± 2.7[d]	<2.5–103	78(16)	2.5[e]	–	<2.5–13.6
V	0.5	77(77)	2.9	3.1 ± 1.6	1.2–12.3	78(78)	9.7	9.7 ± 1.5	4.1–35.2	78(78)	5.7	5.9 ± 1.7	2.2–20.2
Zn	1.3	77(77)	140	126 ± 2.9	12.8–875	78(78)	536	582 ± 2.6	37.4–7420	78(78)	280	288 ± 2.0	70.5–3660

[a] Number in the parenthesis is the number of samples with concentration above the detection limit.
[b] GM = geometric mean, GSD = geometric standard deviation.
[c] Concentrations and detection limit in g/kg.
[d] Geometric mean and standard deviation estimated using Cohen's method.
[e] Median is equal to detection limit because more than 50% of the samples are below detection limit.

often used for statistical analysis of environmental samples (US EPA, 2000). If the number of non-detects exceeded 50% of the samples, then the detection limit is reported as the median, and the geometric mean and standard deviation are not reported.

The concentration of all the metals varied over a wide range. More than 50% of samples of all three fractions contained Ag and Be at concentrations below detection. Se was below detection in about 40% of the fine fraction samples, in more than 40% of the intermediate fraction and in 80% of the paper fraction samples. Pb was below detection in more than 25% of the paper fraction samples.

The mean metal concentrations found in this study were compared with the concentrations reported from other studies. The arithmetic mean was calculated for some specific elements and used for comparison because other studies reported the arithmetic mean of elemental concentrations. The mean (arithmetic) Hg concentrations in the paper, intermediate and the fine fraction were 0.3, 0.6 and 0.3 mg/kg, respectively. The mean Hg concentrations in the intermediate and fine fractions were somewhat higher than the values of 0.38 and 0.13 mg/kg reported by Earle et al. (1999) for the intermediate and the fine fraction, respectively. Flyhammar et al. (1998) investigated concentrations of some metals in the paper and fine fractions of MSW samples collected from a 20 year old test cell in Sweden. The mean (arithmetic) Fe (8316 mg/kg) and Ni (21.7 mg/kg) concentrations in the paper fraction compared well to the 8810 and 21.4 mg/kg reported in the Swedish results, while the mean Mn, Cd, Cr, Cu and Zn concentrations in the paper fraction were found to be much lower than those reported for the Swedish test cell (Flyhammar et al., 1998). The mean (arithmetic) metal content of all metals in the fine fraction were much lower than those reported by Flyhammar et al. (1998). It should be noted, however, that Flyhammar et al. (1998) used 2-mm screen, larger than used in this study (0.425 mm), to separate the fine fraction.

3.3. Comparison between fractions

The fine, intermediate and the paper fractions were compared to each other in terms of elemental concentrations. Because of high variability, the analysis of variance (ANOVA test) could not be used for the comparison of mean metal concentrations of the three MSW fractions (Ott and Longnecker, 2001). The metal concentrations were transformed to their respective natural logarithms. Hartley's test was used to check the equality of the variance of transformed data. If the variances of the transformed concentrations for a metal in all three fractions were found equal, the ANOVA analysis was used to compare the means of the transformed data set. If the null hypothesis of equality of the transformed mean metal concentrations in the three fractions was rejected, Fisher's Least Square Difference (LSD) procedure was used for the comparison of the transformed mean concentrations of that metal in the three fractions (Ott and Longnecker, 2001). ANOVA and Fisher's LSD were used for comparison of mean metal concentrations among the three fractions if the number of non-detects was less than 15%. If the number of non-detects was between 15% and 50%, the geometric means (estimated using Cohen's methods) of the different fractions were compared using the Student's t-test. If the number of non-detects for any fraction exceeded 50%, the Test for Proportions was used to compare the metal concentrations among the different fractions.

Table 2 summarizes the results of the statistical comparisons of the metal concentrations among the three fractions. For each element, the statistical technique performed is identified along with the relative order of each fraction where statistical differences were found. In general, metal concentrations were lowest in the fine fraction and greatest in intermediate fraction. For many of the elements (Al, Ba, Cr, Cu, Co, Fe, Mn, Ni, V and Zn), the mean concentrations in the three fractions were significantly different from each other ($p < 0.001$) and were encountered in the following order: intermediate > paper > fine. The fine fraction, composed mainly of sand, has been reported to have the lowest organic matter content of the three fractions (Miller et al., 1996). The lower organic content of the fine fraction may account for the lower metal concentrations as compared to the other fractions. The mean concentration for all the metals analyzed except As and Ag were lower in the paper fraction than the intermediate fraction. This indicates that the intermediate fraction has greater affinity for the metals than the paper fraction, which also has high organic content. The greater affinity of the intermediate fraction for metals might be because of the smaller particle size providing greater cumulative surface area for interaction.

3.4. Impact of waste age

To evaluate the impact of waste age, the mean metal concentrations of eighteen samples from the three southern most wells (≈8 years old; defined as old samples) were compared with mean metal concentrations of seventeen samples from the three northern most wells (≈3 years old; defined as new samples). Sample ages were estimated from landfill record of waste placement. The southern most (old) samples were also in an area known to be exposed to leachate recirculation in the past. The mean percent paper fraction in the old samples (10%) was significantly ($p < 0.05$) less than that of the new samples (16%). The lower percent of the paper fraction in the old samples as compared to the new ones is likely because of the decomposition of paper over the period

Table 2
Summary of statistical comparisons of metal concentrations among fractions

Element	Statistical comparisons of metal concentrations[a]	Statistical test used
Al	Intermediate > paper > fine	ANOVA
Ag	(a)	Test of proportions
As	Intermediate > fine; paper > fine	ANOVA
Ba	Intermediate > paper > fine	ANOVA
Be	Intermediate > paper; intermediate > fine	Test of proportions
Cd	(a)	Student *t*-tests
Co	Intermediate > paper > fine	Student *t*-tests
Cr	Intermediate > paper > fine	ANOVA
Cu	Intermediate > paper > fine	ANOVA
Fe	Intermediate > paper > fine	ANOVA
Hg	Intermediate > paper; intermediate > fine	ANOVA
Mn	Intermediate > paper > fine	ANOVA
Ni	Intermediate > paper > fine	ANOVA
Pb	Intermediate > fine > paper	Student *t*-tests
Se	Intermediate > paper; intermediate > fine	Test of proportions
V	Intermediate > paper > fine	ANOVA
Zn	Intermediate > paper > fine	ANOVA

(a) No statistical difference among fractions.
[a] Statistical significance tested at 99.9% confidence.

of time. In general, the metal concentrations in all the three fractions of old samples were greater than those of new samples. The mean Al, Fe, and Cr concentrations in all three fractions of the old MSW samples were significantly ($p < 0.05$) greater than those of the new samples (Table 3). The mean Fe concentrations of all the three fractions of the old samples were ≈3 times that of the new ones. The mean Al concentrations of all the three fractions of the intermediate fraction of the old samples were ≈4 times that of the new ones. The mean Ba, Cd, and Cu concentrations in the fine and paper fractions of the old MSW samples were significantly ($p < 0.05$) greater than the new samples. The mean As concentrations in the fine and intermediate fractions of the old sample were significantly greater than the new samples.

In order to explore the effect of sample age on the elemental content of the parent MSW samples, metal concentrations of the composite samples were calculated. The fine, intermediate and paper fractions represented the entire sample taken from the landfill minus the oversized non-biodegradable and very slowly biodegradable material. The metal concentrations of the composite (paper + intermediate + fine) samples were determined by multiplying the percentage of the total sample weight that each fraction represented by the metal concentration of that fraction, adding the resulting three numbers together and the dividing by the percentage of total

Table 3
Metal concentration in old and new MSW samples (mg/kg dry basis)

	Al[a]	As	Ba	Cd	Cr	Cu	Fe[a]	Pb	Mn	Hg	Ni	V	Zn
Fine arithmetic mean													
Old	12.4a	2.4a	26.3a[b]	1.2a[c]	18.2a	44.3a	7.7a	29.2a	135	0.50	9.0	4.0	253
New	3.9b	1.4b	17.4b	0.4b[c]	9.9b	17.8b	2.6b	10.1b	90.3	0.17	25.2	3.2	175
Intermediate arithmetic mean													
Old	36.1a	6.5a	65.4	1.7[c]	86.4a[b]	225	28.1a	51.7	644	1.44a	26.2	10.5	861
New	8.7b	3.8b	68.6	2.4	48.8b	308	10.1b	38.3	988	0.28b	65.3	9.1	898
Paper arithmetic mean													
Old	14.7a	11.9	56.6a	2.1a	27.2a[b]	69.3a[b]	12.3a	37.5	179a	0.40a[b]	11.8	8.4a	314
New	5.7b	6.2	32.7b	0.9b	39.3b	47.4b	4.5b	18.0	67.8b	0.21b	45.4	5.1b	345
Composite arithmetic mean													
Old	16.2a	7.9a[b]	54.5	2.3	63.3	174	18.8a	46.2	451a	0.92a	20.1	9.3a	662
New	4.9b	4.4b	55.0	1.7	42.3	197	6.8b	34.8	377b	0.21b	50.5	7.6b	690

Mean concentrations within a column (for a fraction) followed by different letter are significantly different ($p < 0.05$).
[a] Concentration is in g/kg dry basis.
[b] Significance level $p < 0.1$.
[c] Mean estimated using Cohen's method.

sample weight that the three fractions represented together (Eq. (1)).

$$\text{Concentration of metal } M_j \text{ in composite sample} = \frac{\sum_i C_{ij} p_i}{\sum_i p_i}, \quad (1)$$

where i is the paper, intermediate and the fine; p_i, the percentage of ith fraction in total sample weight; C_{ij}, concentration of metal M_j in ith fraction.

The mean Al, As, Fe, Mn, Hg, and V concentrations in the old composite samples were found to be significantly ($p < 0.05$) greater than those in the new samples. The lower Hg content of the new samples could be the consequence of a sharp decline in the disposal of Hg batteries in MSW, the major source of Hg in the MSW stream (US EPA, 1997b), from 29.3 metric tons in 1989 to 6.7 metric tons in 1995 in Florida (FDEP, 1996). The increased concentration that could be caused by the extended degradation of organic for old samples compared with new samples would not be great enough to be the sole cause of the increased concentrations seen. The higher concentrations of many of the other metals in the old MSW samples as compared to the new samples are most likely because of redistribution of these metals as a result of leaching and waste decomposition.

3.5. Management of reclaimed material

The feasibility of a landfill reclamation project will largely depend on management of the reclaimed material. As mentioned earlier, the cover soil and degraded organic waste have been reported to constitute ≈75% of the recovered material and possible reuse options for the recovered soil include landfill cover material and construction fill (US EPA, 1997a). Before such material can be recycled outside of the landfill, the risk posed by elevated pollutant concentrations should be evaluated. An examination of such risk is usually required by regulatory agencies and elevated pollutant concentrations may limit reuse options. Possible limitations to the land application of reclaimed soils from the Florida landfill under study were evaluated by comparing the concentrations of the combined fine and intermediate fractions (denoted herein as "residual soil") to the guidelines or regulations of several different environmental agencies.

The residual soil concentrations were obtained using Eq. (1) for the intermediate and fine fractions only. One-half of the detection limit was used when concentrations were below detection. Sufficient data existed to calculate the metal concentrations in the residual soil for a total of 71 samples.

The arithmetic mean was calculated for some specific elements and used for comparison with data reported by Das et al. (2002), who reported the mean metal concentrations of six composite samples from a Georgia, US, landfill; these samples would have been comparable to the residual soil fraction reported here. The concentrations of As, Cd, Cu, Pb and Zn in the residual soils from the Florida landfill were found to be comparable to the values reported for the Georgia landfill, while the mean Ni and Cr concentrations (17.2 and 27.4 mg/kg, respectively) were found to be much lower in the Florida samples.

Upper 95% confidence limits (UCL) of the metal concentrations assuming a log normal distribution were compared to regulatory thresholds for the US, the UK and the Netherlands. The 95% upper confidence level is often used when comparing variable field results to regulatory thresholds. Table 4 provides for each metal the number of samples, the geometric mean and standard deviation, the 95% log-normal UCL and the concentration range, along with the regulatory thresholds.

As Table 4 illustrates, regulatory thresholds can vary dramatically for some elements. The US EPA developed soil screening levels (SSL) as part of their soil screening guidance (SSG) for cleanup of contaminated sites (US EPA, 1996). Many US states have adopted a similar risk assessment methodology to develop their thresholds for clean soils and land-applied wastes. Similar risk-based soil thresholds have been developed by a number of European countries. Included in this comparison are soil guideline values (SGV) developed by the United Kingdom Department of Environment, Food and Rural Affairs and Environment Agency and intervention values (IV) developed by the Netherlands Ministry of Housing, Spatial Planning and Environment (MHSPE). Differences in threshold values for a given element result from differences in assumptions on toxicity, acceptable risk, exposure scenarios, and algorithms for calculating thresholds.

When the 95% UCL values were compared to the different thresholds, three different elements appear to pose the greatest concern with respect to possible limitations: As, Be, and Cd. Arsenic concentrations in 67 samples out of 69 exceeded the US EPA SSL (0.4 mg/kg). Arsenic did not exceed the UK SGV or the Netherlands IV for arsenic, as these thresholds are much larger than the US EPA SSL. The US EPA SSL was developed for arsenic as a carcinogen and determined at a 1 in 1,000,000 cancer risk. The Florida Department of Environmental Protection's soil cleanup target level for arsenic in residential settings (developed using methodology and assumptions similar to the SSLs) is 0.8 mg/kg (FAC, 1999). Arsenic concentrations would likely limit the reuse of these residual soils in Florida to residential areas. When Das et al. (2002) assessed the metal concentrations in soils from the Georgia landfill for land application, they compared the results (which were similar to this study) to standards promulgated for the land application of biosolids and con-

Table 4
Comparison of metal concentrations in residual soil with risk-based concentrations[a]

	Number of samples	Metal concentration in residual soil (mg/kg dry basis)			Risk-based metal concentrations in soil (mg/kg)[b]		
		GM ± GSD	95% Upper confidence level for mean	Range	SSL[c] (mg/kg)	SGV[d]	IV[e] (mg/kg)
Al[f]	71	6.6 ± 0.001	7.6	2.4–73			
As	69	2.0 ± 0.7	2.4	0.3–13.7	0.4(67)	20	55
Ba	71	26.2 ± 0.4	29.9	9.6–356	5500		625
Be	71	0.1 ± 0.3	0.1	<0.1–0.5	0.1(35)		
Cd	71	0.9 ± 1.2	1.1	<0.3–21.2	78	1(31)	12(1)
Co	71	3.5 ± 0.6	4.0	<1.8–430			240
Cr	71	17.7 ± 0.7	20.9	3.2–331	390	130(1)	380(1)
Cu	71	38.7 ± 1.3	48.4	3.3–738			190(5)
Fe[f]	71	5.8 ± 0.001	6.9	1.0–42.6			
Pb	71	15.3 ± 1.7	19.9	<2.0–403	400(1)	450	530
Mn	71	80.6 ± 1.1	99.1	9.7–2510			
Hg	69	0.2 ± 1.2	0.2	0.05–4.7	23		10
Ni	71	9.1 ± 0.9	11.0	1.6–369	1600	50(1)	210(1)
Se	71	3.7 ± 0.4	4.1	<2.5–69	390	35(1)	
Ag	71	1.1 ± 0.5	1.3	<1.5–20.6	390		
V	71	4.6 ± 0.3	5.1	1.7–22.8	550		
Zn	71	238 ± 1.3	247	22–7420	23,000		720(9)

[a] Residual soil is composed of the fine and retained fraction.
[b] Number is the parenthesis is the number of samples that exceeded the respective risk-based concentration.
[c] US EPA soil screening levels corresponding to ingestion pathway.
[d] UK EA soil guideline values corresponding to sandy soil, residential (with plant uptake) land-use. The SGV for Cd is a function of soil pH (value for pH 6.0 is shown).
[e] Netherlands MHSPE intervention value.
[f] Concentrations in g/kg.

cluded land application would be appropriate. The arsenic standard determined in the biosolids rule does not consider arsenic carcinogenicity, while most state guidelines do. This illustrates the importance of utilizing the appropriate thresholds for comparison.

While 19 samples of the 71 samples exceeded the US SSL for Be (0.1 mg/kg), it should be noted that ≈80% of the fine and 50% the of intermediate fraction samples were below detection and the detection limit of Be was equal to the SSL. Additional work at lower limits of detection would therefore be recommended to assess true risks posed by Be. No UK soil guidelines levels or Netherlands intervention values were available for Be. Cd was found to be a concern with respect to the UK SGVs. Since plant uptake is factored into the development of the SGV, assumed soil pH has an impact of the SGV used for comparison. Table 3 shows an SGV for cadmium of 1 mg/kg which corresponds to a soil pH of 6.0. At a soil pH of 7, the SGV is 2 mg/kg. Only one sample exceeded the Netherlands IV for Cd, while no samples exceeded the US SSL for Cd.

Since arsenic was the most limiting element with respect to reuse (considering the US SSL), an effort was made to identify the source of As in the residual soil. As indicated in Table 1, the geometric mean of As concentration of the fine fraction, which is primarily cover soil and which represented approximately half of sample mass and ≈75% of the "residual soil" fraction, was 1.4 mg/kg. The arsenic concentrations in the intermediate and paper were fraction were larger than this. Samples of natural soil, which were believed to be representative of the cover soil used during landfill operation, were collected and analyzed for arsenic. The arsenic concentration in each of eleven composite soil samples collected was below detection (0.2 mg/kg). Thus the source of the arsenic appears to be the solid waste itself. Apparently during the process of mixing soil with waste in the landfill, and the subsequent leaching and decomposition occurring, arsenic concentrations in the fine fraction (the cover soil) became elevated. The exact source of the arsenic and the mechanism of transport to the fine fraction are unknown. This merits further research.

4. Conclusions

The objectives of this study were to characterize soil and fine organic matter recovered from a MSW landfill with respect to the metal content and to assess limitations associated with the reuse of these materials outside the landfill environment. Samples were collected from a MSW landfill in Florida and separated into different size fractions. Approximately 45% of the reclaimed material (by weight) was composed of the

sand that was used as daily and intermediate cover material. Another 15% was composed of stabilized organic material (>0.425 and <6.3 mm). The paper fraction constituted about 12% of the recovered material. In general, the metal concentrations were greatest in intermediate fraction and least in the fine (soil) fraction. The mean Al, Ba, Cr, Cu, Co, Fe, Mn, Ni, V, and Zn concentrations in the three fractions were significantly different from each other ($p < 0.001$) and were encountered in the following order: intermediate > paper > fine. The effect of age on metal concentrations was found to be dependent on the metal and the fraction. The mean Al, Fe, and Cr concentrations in all three fractions of the old MSW samples were significantly ($p < 0.05$) greater than those of the new samples. The mean Al, As, Fe, Mn, Hg and V concentrations in the old composite samples were found to be significantly ($p < 0.05$) greater than those in the new samples. Lower Hg concentrations in the new samples may be the consequence of a sharp decline in the amount of Hg in household products disposed in MSW. The higher Al and Fe concentrations of the old MSW samples as compared to the new samples are most likely because of redistribution of these metals as a result of leaching and waste decomposition.

To assess the possible limitations of reuse of the reclaimed soil (consisting of fine and intermediate fractions) outside of the landfill, metal concentrations were compared to regulatory thresholds from the US, UK and the Netherlands. For the most part, metal concentrations were below the regulatory thresholds, though As, Be and Cd may be a concern. Elevated concentrations of As presented the most concern with respect to reuse when compared to US thresholds that treat arsenic as a carcinogen; arsenic was a not a problem when compared to the European thresholds. The source of the arsenic was the waste itself, and not the cover soil. Cadmium was not a limiting element using the US thresholds, but was limited under the UK thresholds. Many assumptions go into the development of risk-based regulatory thresholds, and they are oftentimes conservative in nature. A more detailed site-specific study to evaluate the risk associated with the land application of the residual soil reclaimed from an MSW landfill would be beneficial. The impacts of pollutant leaching or the impacts of organic chemicals were not evaluated and may also have to be addressed prior to a reclamation project.

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Solid waste from aluminum recycling process: characterization and reuse of its economically valuable constituents

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Abstract

Due to economic advantages, many companies in Brazil recover Al from the process of crushing and water-leaching of secondary aluminum dross. Wastes from this process (non-metallic products and salts) are usually landfilled or disposed without treatment, causing many environmental damages. The purpose of this work is to investigate, in a recycling company sited in São Paulo metropolitan area (Brazil), the potential use of the non-metallic product (NMP) in the production of concrete blocks and to evaluate the presence of important chemical compounds that may be useful for other applications. Chemical and mineralogical analyses revealed that NMP is composed of refractory and abrasive oxides (α-Al_2O_3, $MgAl_2O_4$, SiO_2) and an important source of transition alumina: α-$Al(OH)_3$. Concrete blocks were made by adding two parts of NMP to one part of cement and four parts of sand. The blocks were tested according to the Brazilian standard (NBR7173/1982) and they passed dimension, humidity and absorption tests but not compressive strength tests. However, particular NMP constituents have accelerated the strength rate development of the blocks, thus decreasing working time. The commercial use of NMP can reduce the amount of discarded wastes contributing to environmental preservation.
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1. Introduction

Aluminum production starts by refining bauxite through the Bayer process, from which alumina is obtained. From the Hall-Héroult process, the alumina is electrochemically processed and metallic aluminum is produced. Due to the metal capacity of becoming oxidized during its fusion, even in the presence of cryolite (Na_3AlF_6), a large amount of dross is generated. Dross resulting from this process usually contains about 80% (wt) of metallic aluminum (Carvalho, 1991), which can be recycled by secondary industries. These industries also recycle any type of aluminum scrap, such as aluminum house siding, castings and used beverage cans. Besides the metallic content, other valuable chemical compounds can be recovered from the process. For example, Rabah (2003) updated the current recycling technology by adding the removal of the coating paint and decreasing magnesium losses during the melting process of used beverage cans, to recover standard aluminum–magnesium alloys and some valuable salts.

Recycling is an economical and environmentally friendly way to handle some types of hazardous wastes, reducing the amounts disposed in landfills.

In Brazil, research on industrial waste recycling started to become popular in the 1970s, with the increase of the steel-making sector, which generates approximately four million tons of dross per year (Instituto de Pesquisas Tecnológicas do Estado de São Paulo, 1994). In the early 1990s, with the introduction of aluminum beverage cans in the national market, aluminum recycling activities started to grow. Since then the sector has continuously grown and today Brazil is one of the world's largest aluminum recyclers, ahead of Japan

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and the United States (Associação Brasileira do Alumínio, 2004).

1.1. Aluminum dross recovery process

Because of the high metallic aluminum content (about 80% (wt)) dross produced by the primary aluminum industry is known as white dross (Carvalho, 1991). Low-quality dross produced by the secondary aluminum industry containing around 5–20% Al and a large quantity of soluble salts, is usually known as black dross, and those containing between 5% and 10% Al are known as salt cake (Gwinner, 1996). Almost 800,000 ton of salt cake is annually landfilled in the United States and this number continues to grow with the increase in aluminum consumption, mainly recycled metal (Sreenivasarao et al., 1997). In Europe the landfill disposal of this waste is forbidden because the slag soluble salts represent a potential source of pollution to surface and groundwater supplies (Gwinner, 1996).

In order to solve the problem of salt cake disposal, many technologies have been tested or proposed. Some of them involve water leaching of salts from salt cake at high temperatures and pressures (Bodnar et al., 1997), the use of plasma arc and low oxygen furnaces (Schirk, 1997) or even eletrodialysis for the recovery of salts from leaching solutions (Sreenivasarao et al., 1997). These alternative techniques are not economically feasible and do not maximize aluminum recovery; they also produce aluminum oxide and other residues, which require landfill disposal.

The first submitted methodology for aluminum dross recovery was patented in the 1970s in United States by Papafingos and Lance (1978). This patent features equipment for cooling and disaggregating aluminum dross with water in order to dissolve the salts. During the aluminum dross digestion, several undesirable and potentially toxic chemical reactions end up producing hydrogen, methane and ammonia gases.

According to Yerushalmi (1992), to prevent those undesirable reactions, pH in the digester must be maintained below 8, and preferably above 5 through magnesium chloride addition, which can be taken from a crystallizer that recovers the salts from the process itself. This salt suppresses the reactions that increase the leaching liquid pH since it reacts with the hydroxyl ions in alkali solutions to form non-dissociated $Mg(OH)_2$ and HCl. HCl decreases the pH and consequently slows down the AlN and the metallic aluminum reactions with water. In order to avoid contamination in the recovered salt flux (NaCl/KCl) with the addition of $MgCl_2$,the method developed by Yerushalmi (1992) recycles magnesium from aluminum dross without increasing the overall amount of $MgCl_2$ in the system.

In addition to the recovery of metallic aluminum, salt flux and magnesium chloride, the process generates a waste, known as non-metallic (NMP), which is usually disposed in landfills.

NMP can be useful as an exothermic topping in steel manufacture, because of its heat-generation and retention properties. These properties result from its high content of metallic aluminum fines as well as aluminum oxide.

Yerushalmi and Sarko (1995) proposed a method where NMP undergoes several reactions enabling its inorganic constituents to be joined to a ceramic matrix. To obtain a ceramic abrasive or residual material, they determined that the waste must show around 40% (wt) to 75% (wt) of Al_2O_3, 5% (wt) to 20% (wt) of MgO and 2% (wt) to 15% (wt) of SiO_2; the remainder may be composed of oxides of Ti, Cu, Zn, Ca, Na and other trace elements. The remainder may also contain nearly 2% (wt) of Cl. Another interesting method for recovering valuable products from the NMP was introduced by Pickens and Waite (2000) where alumina hydrate, magnesium hydroxide and magnesium aluminate are separately recovered, thereby increasing their commercial values. This process treats the NMP in an acid digester to selectively dissolve alumina and magnesia, while the undissolved magnesium aluminate is separated by filtration. In order to recover the dissolved constituents, the pH solution is raised to about 9.5–12 to precipitate magnesium oxide, which is removed by filtering. With a pH of about 10–11, the aluminum is held in solution and the mixed oxides remain as solids that are also removed by filtration. When the pH of the remaining liquid nears neutrality, alumina trihydrate precipitates and the result is a substantially pure product. According to the authors, this process consumes significant amounts of acids and bases, which can be regenerated, for example, by selective membrane technology. Pickens and Morris (2001) presented a process where a non-metallic product, substantially free of metallic aluminum and aluminum nitride, can be used to produce calcium aluminate by mixing NMP with calcium oxide. After the heating stage, the final product (calcium aluminate) is used to remove undesirable elements, such as sulfur, in high-quality steel.

Fine grains (less than 150 μm) of the NMP can also be used as raw material in cellular concrete (Araújo, 2002). The aluminum content in this fraction reacts with water during the production of the concrete while releasing hydrogen. This reaction promotes the formation of pores that reduce concrete density without affecting its strength.

The method proposed by Rabah (2003) recovers some valuable salts (salts of aluminum, magnesium and tin) from the slags produced by the melting process of used beverage cans with a mix of sodium borate-sodium chloride as a salt flux.

Many secondary recycling industries in Brazil recover metallic aluminum using a simple process of crushing

and melting of white dross in the presence of a salt flux (NaCl/KCl). Therefore, new dross is generated (black dross and salt cake), which are reused as raw material by tertiary aluminum industries (Fig. 1). Although in the USA and in many European countries only white dross can be recycled and the black dross is landfilled (when it is permitted), in Brazil many secondary recycling industries supply tertiary companies with their slags (for free or at a lower cost).

Therefore, the activities of tertiary companies represent an advantageous way for the primary and secondary industries to solve the waste disposal problem.

The purpose of this work is to investigate the potential use of the non-metallic product (NMP) from a tertiary aluminum company producing concrete blocks. The company is sited in São Paulo metropolitan area (São Paulo State, Brazil). The work also evaluates the use of its chemical constituents in other applications. These investigations are based on the chemical and mineralogical characterizations of the wastes under study.

2. Recycling process of a tertiary aluminum industry – case study

Many tertiary aluminum-recycling companies in the São Paulo metropolitan area operate using a simple method for treating black dross. In this process the dross is crushed using a vertical hammer mill to release the metallic portion. The remaining material is water leached in a rotary drum (a rotating barrel of about 5-m length and 1.90-m diameter). The recovered material is sorted by size using a 20-mesh sieve. Particles larger than 20 mesh (containing about 60% (wt) to 80% (wt) of Al) are sent to secondary industries for remelting. Particles smaller than 20 mesh, which represent 25% (wt) of Al can be sold to steel manufacturers as an exothermic product.

During the dross leaching process a semi-liquid waste, rich in soluble salts, is produced, which is disposed in tanks for decanting the solid fraction. The measured pH of the leaching solution in those tanks is about 9.

The decanted solid material, termed non-metallic product (NMP), has a low content of Al and is eventually disposed in landfills. Due to the high cost of treatment and disposal, it is frequently disposed without any previous treatment in the environment (for example, in ponds and rivers).

Brazilian regulations (Conselho Nacional do Meio Ambiente, 1986) consider wastes from the tertiary aluminum industry as "non-hazardous" materials. However, this designation is modified if toxic and inflammable gases such as ammonia, hydrogen and methane are present. According to Filleti (1995), ammonia is produced from the hydrolysis of nitrides (Eq. (1)), which are generated during the melting process by the reaction of atmospheric nitrogen with aluminum metal. Hydrogen is generated from the hydrolysis of metallic aluminum (Eq. (2)) (Araújo, 2002) and methane by the reaction of carbides with water (Eq. (3)). Carbides are produced from the oils of aluminum scraps during the melting process (Shinzato, 1999).

$$2AlN + 6H_2O \rightarrow 2Al(OH)_3 + 2NH_3 \uparrow \quad (1)$$

$$2Al + 2H_2O + 4OH^- \rightarrow 2Al(OH)_3 + H_2 \uparrow \quad (2)$$

$$Al_4C_3 + 12H_2O \rightarrow 4Al(OH)_3 + 3CH_4 \uparrow \quad (3)$$

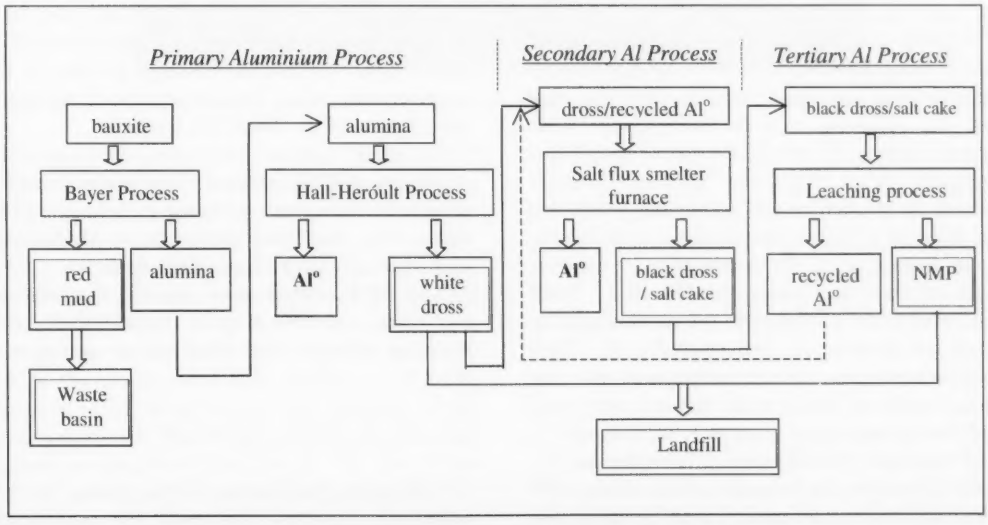

Fig. 1. Aluminum schematic recycling process in Brazil.

The tertiary aluminum company evaluated is capable of treating 20 ton of black dross per day, recovering 4 ton of Al and 16 ton of solid waste, which has no commercial value. In 2000 the unit price of black dross in the São Paulo metropolitan area was around US$7/ton, and recovered Al was US$133/ton. Because of economic advantages, this activity is increasing in large cities where consumption of aluminum materials is higher than in other places.

In 2000 the price charged by industrial landfills in the region, was about US$20/ton of waste.

3. Methodology

In this study the following materials from the tertiary aluminum company were chemically and mineralogically characterized:

1. *Dross* – (*black dross*) used as raw material by the tertiary company and produced from the melting process of white dross and aluminum scrap by a local secondary industry.
2. *NMP* (*non-metallic product*) – solid wastes decanted in tanks, a result of the water leaching process of the black dross.
3. *Crust* – hard solid material produced by continuous leaching of NMP and precipitation of aluminum hydroxides, usually developed inside pipings where the wastes from the decantation tanks are disposed into a pond. Due to this formation, the piping is sometimes clogged.

The studied samples (dross, NMP and crust) were dried (54 °C) and crushed (200 mesh) before being analyzed. To determine chemical constituents, these materials were analyzed by X-ray fluorescence method (Philips/PW2400) and the mineralogical composition was obtained using Siemens D5000 X-ray diffractometer. The black dross and crust samples were also analyzed with the scanning electron microscopes (Leo/440i) equipped with Si–Li energy dispersive spectrometer (Oxford).

To evaluate an alternative way of reusing NMP, and thus avoid disposal in the environment, the material was tested as an aggregate in the production of concrete blocks without structural function. Therefore, NMP was washed with water to eliminate the remaining soluble salts and gas production, and dried (54 °C). Then this material was sent to a local manufacturer that usually produces concrete blocks using the following proportion of cement/aggregate: one part of cement for six parts of aggregate (usually sand). Since the proportion actually decreases the strength of the blocks, this product has no structural function and by being less expensive it is affordable to the people of the region. In this study two parts of natural aggregate were replaced by NMP, which resulted in the following proportion: 1 part of cement:2 parts of NMP:4 parts of natural aggregate. Enough water was added to this mixture for obtaining a workable mortar, facilitating the molding of the blocks. The produced concrete blocks were kept at ambient temperature until they could be transported and stored in another place – this period is known as "working time".

Finally, the characteristics of the blocks produced with NMP (about 30 units, aged 28 days) were analyzed using conventional physical-mechanical tests (dimensional, humidity, absorption and compressive strength) recommended by Brazilian standard NBR 7173/1982 (Associação Brasileira de Normas Técnicas, 1982). The dimensional test verifies whether the length, height and width of the blocks are in accordance with the standard measures. The humidity test investigates the hygroscopic properties of the blocks, the absorption test checks their impermeability. The compression test determines the capacity of the blocks to withstand a specific load, checking if these materials offer adequate mechanical strength.

4. Results

4.1. Chemical and mineralogical characterizations

According to the results of the chemical analysis (Table 1), over 80% (wt) of the dross is composed of soluble salts constituents (such as Na, K and Cl), the rest is made up of aluminum and scraps alloying elements (Mg, Si, Fe, Ca, Ti, Mn, etc.). NMP and the crust are mainly comprised of Al, Si, Mg and Fe.

XRD investigations revealed that aluminum dross is constituted of NaCl, $MgAl_2O_4$, Al_2O_3 and SiO_2, while the NMP consists of the same oxides plus $Al(OH)_3$ (Table 2). Since an intense leaching process of the waste generates the crust, its composition is different due to the absence of the $MgAl_2O_4$ (spinel).

Scanning electron microscopy investigations of the dross revealed the presence of many microscopic spheres of metallic aluminum occluded in the inside of the oxide plates (Fig. 2). These oxides are α-Al_2O_3 (corundum) and $MgAl_2O_4$ (spinel) and SiO_2 (quartz).

The SEM analysis crust showed that the aluminum hydroxide (needle-shaped) precipitated over the rounded alumina and silica grains acting as cement (Fig. 3).

4.2. Concrete blocks with NMP

During the production of the blocks, working time only took one day in this study; this usually takes four days in the manufacturer's process.

Table 1
Dross, NMP and crust chemical composition

(wt%)	Na_2O	Al_2O_3	MgO	SiO_2	K_2O	CaO	Fe_2O_3	MnO	P_2O_5	TiO_2	BaO	Cr_2O_3	NiO	CuO	PbO	ZnO	Cl
Dross	34.65	11.11	2.12	2.23	0.18	0.33	0.80	0.05	0.06	0.19	0.02	0.01	0.00	0.02	0.01	0.00	47.96
NMP	1.69	68.18	9.73	13.04	0.52	0.82	1.80	0.32	0.11	0.61	0.04	0.10	0.01	0.08	0.01	0.03	2.00
Crust	0.60	69.91	4.33	20.93	0.38	0.46	1.97	0.51	0.09	0.34	0.02	0.10	0.01	0.09	0.00	0.03	0.22

Table 2
Dross, NMP and crust mineralogical phases

Sample	Mineralogical phases			
Dross	NaCl (*halite*)	$MgAl_2O_4$ (*spinel*)	Al_2O_3 (*corundum*)	SiO_2 (*quartz*)
NMP	$MgAl_2O_4$ (*spinel*)	Al_2O_3 (*corundum*)	$Al(OH)_3$ (*bayerite*)	SiO_2 (*quartz*)
Crust	Al_2O_3 (*corundum*)	$Al(OH)_3$ (*bayerite*)	SiO_2 (*quartz*)	

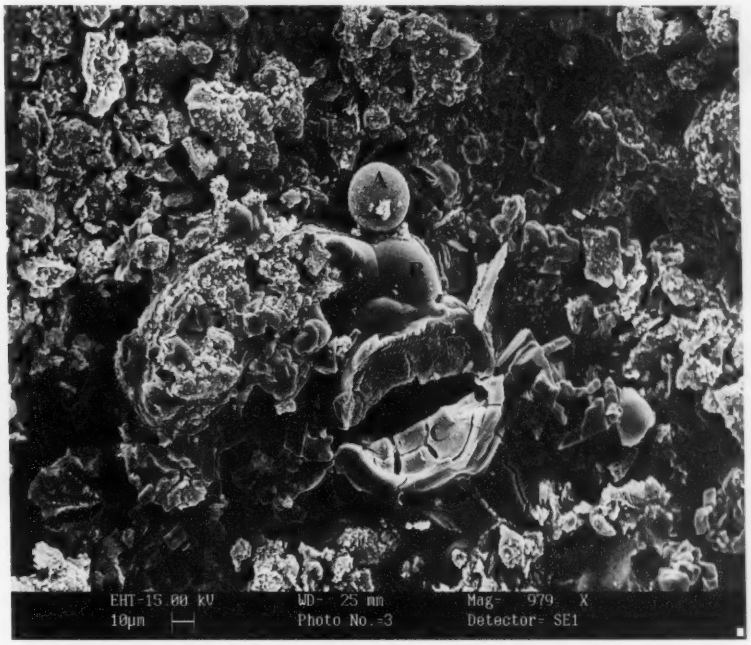

Fig. 2. Dross SEM images: (A) metallic aluminum sphere; (B) metallic aluminum sphere occluded inside the oxide plates; (C) oxides plates.

The texture of the manufactured blocks revealed a flat, homogeneous and compact surface. They did not show any fracture or other surface failures.

According to Brazilian standards, twelve units of concrete blocks with NMP were submitted to the tests to verify their quality. Humidity and absorption tests were carried out in samples 1–6, while samples 7–12 were used in the compression tests (Table 3).

Dimensions of all tested blocks are accurate within the specific standards. The samples of blocks also met the standards of the humidity tests. In the absorption test only the individual values were adequate. The results obtained in the compressive strength tests were not satisfactory; nevertheless, sample No. 11 almost reached the value established by the standard for the individual mean (2.0 MPa).

5. Discussion

5.1. Chemical and mineralogical characterizations

Since NMP represents the product of the leaching process of the aluminum dross, its composition is mainly comprised of the insoluble constituents, such as aluminum oxide, silicon oxide and magnesium oxide. On the other hand, the crust represents the leached product of

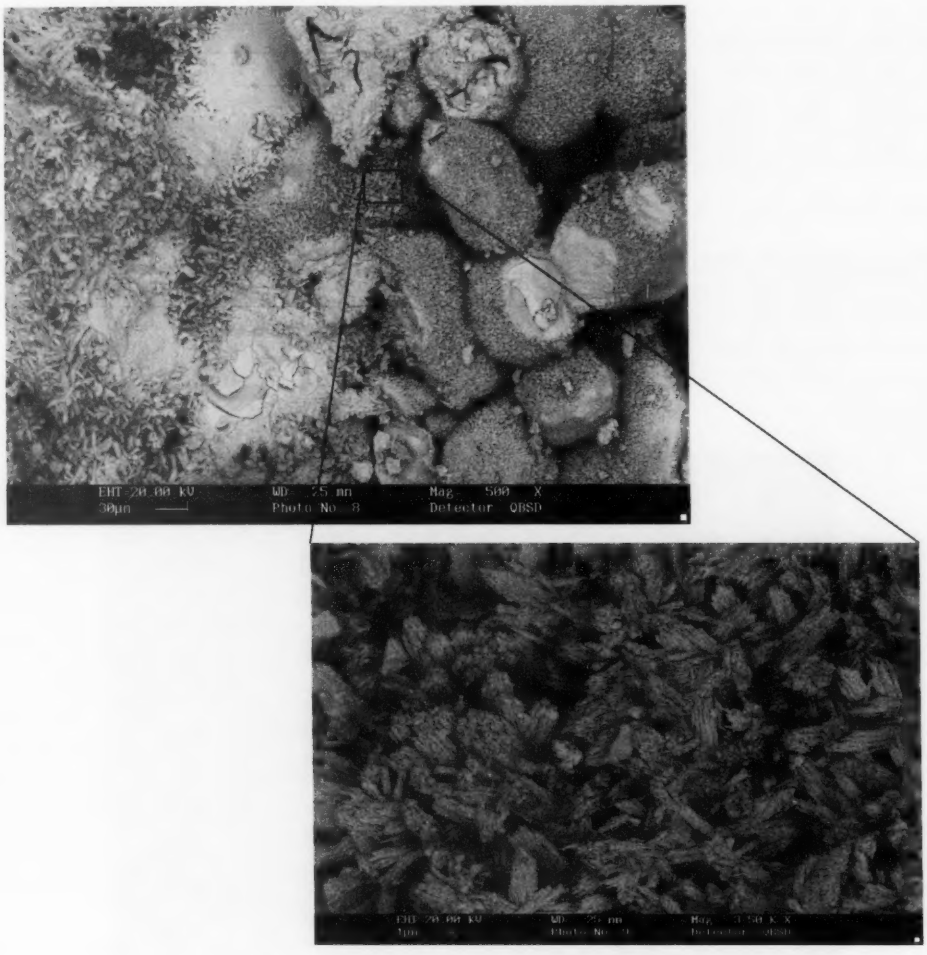

Fig. 3. Crust SEM images. Bayerite particles in detail.

Table 3
Results of dimensional (length, height and width), humidity, absorption and compressive strength tests on blocks with NMP

Block sample	Length (mm)	Height (mm)	Width (mm)	Humidity (%)	Absorption (%)	Compressive Strength (MPa)
1	391	189	140	14.8	11.1	
2	390	186	141	11.7	13.2	
3	391	188	141	20.6	10.3	
4	390	189	141	15.8	10.2	
5	391	188	141	14.8	10.0	
6	390	188	141	12.4	12.1	
7	391	188	140			1.4
8	391	189	140			1.2
9	391	188	140			1.3
10	391	189	141			1.5
11	390	189	141			1.8
12	390	189	140			1.1
Mean				15.02	11.15	1.4
Limit according to NBR 7173/1982	390 mm (+3 and −2 mm)	190 mm (+3 and −2 mm)	140 mm (+3 and −2 mm)	Mean < 40%	Mean < 10% Individual < 15%	Mean > 2.5 MPa Individual > 2.0 MPa

NMP. For this reason, the concentration of soluble salts in this material has decreased.

In order to understand the particular geochemical behavior during the recycling process of some elements present in dross, NMP and crust, the relative removal or accumulation values of these elements were determined. Since dross leaching produces NMP and crust, only the major constituents of the dross were taken in consideration (with concentrations over 0.1% (wt)). The constituents for this case are NaO, Al_2O_3, SiO_2, MgO, Fe_3O_2, K_2O, CaO, TiO_2 and Cl. If all these compounds were considered as a molar group ($n_1 + n_2 + \cdots + n_9$), where n_1 is the number of moles of NaO, n_2 the number of moles of Al_2O_3 etc., then each of them would correspond to a fraction of the group. Thus, in a group of nine compounds, the molar fraction of one element (X_i) can be obtained through the following expression:

$$X_i = \frac{n_i}{n_1 + n_2 + \cdots + n_9},$$

where $0 \leqslant X_i \leqslant 1$ and $\sum X_i = 1$.

The chemical concentration values of the nine compounds were normalized and molar fraction variations (ΔX) between NMP and dross and also those between crust and NMP were determined to verify the relative removal or accumulation of elements in the final products (Table 4). The positive values mean accumulation and the negative ones mean removal (Hypolito, 1972).

The results of NMP ΔX (Table 4 – column 5a) indicate a substantial removal of Na and Cl during the leaching process and relative accumulation of Al, Mg, Si, Ca and Fe. The ΔX values of crust (Table 4 – column 5b) reveal the removal of Mg, Cl, Na, Ca, Ti and K during the NMP leaching process and a consequent accumulation of Si, Al and Fe.

The oxides present in the NMP and crust are produced during the melting process that originated the dross. The corundum (also known as α-alumina) comes from the reaction of the atmospheric oxygen with metallic aluminum molten at high temperatures. This reaction is exothermic (Eq. (4)) and can also raise the local temperature to 2000 °C, thus increasing the production of dross (Filleti, 1993).

$$2Al + \tfrac{2}{3}O_2 \rightarrow Al_2O_3 + 399 \text{ kcal/mol} \quad (4)$$

$MgAl_2O_4$ and SiO_2 are the result of oxidation of magnesium and silicon present in the aluminum scrap as alloying elements, during the metal melting. According to Carvalho (1991) for each 1% of Mg present in the metal, 10–15% of $MgAl_2O_3$ will be generated, thus increasing the production of the dross.

The α-alumina is the main oxide present in the studied wastes. It has a hexagonal structure in which all of the aluminum atoms are octahedrally coordinated. This oxide is very stable and has a very high melting point (2050 °C), extreme hardness (9 on the Mohs scale), high electrical resistivity (10^{11} Ωm at 500 °C), low dielectric constant and high thermal conductivity (Moulson and Hebert, 1990). The main applications of α-Al_2O_3 are manufacturing of bricks, glass and enamel, tiles and porcelain, mechanical, electric and electronic ceramics, etc. (Ruberto, 1998).

Studies are showing the potential use of non-metallic wastes as refractory and abrasive materiai. The chemical composition of NMP and the crust are in agreement with those established by Yerushalmi and Sarko (1995) for wastes with abrasive clay, i.e.: 40% (wt) to 75% (wt) of Al_2O_3, 5% (wt) to 20% (wt) of MgO and 2% (wt) to 15% (wt) of SiO_2; 2% (wt) of Cl.

Microstructural analyses showed that dross is composed of oxide plates that retain the liquid aluminum (Fig. 2). According to Tenório et al. (1995) the plate shape of oxides is due to the action of salt used as a flux to break the chain structure. This way the aluminum is released from the oxide structure, and tends to coalesce

Table 4
Chemical concentration (1) of major components of dross, NMP and crust samples and their corresponding normalized values (2)

	Dross				NMP					Crust				
	(1) wt%	(2) wt%	(3) n	(4) X	(1) wt%	(2) wt%	(3) n	(4) X	(5a) ΔX	(1) wt%	(2) wt%	(3) n	(4) X	(5b) ΔX
Na_2O	34.650	34.800	0.561	0.263	1.690	1.718	0.028	0.022	−24.118	0.600	0.605	0.010	0.008	−1.365
Al_2O_3	11.110	11.158	0.109	0.051	68.180	69.296	0.680	0.535	+48.375	69.910	70.516	0.692	0.578	+4.304
MgO	2.120	2.129	0.053	0.025	9.730	9.889	0.245	0.193	+16.841	4.330	4.368	0.108	0.091	−10.258
SiO_2	2.230	2.240	0.037	0.017	13.040	13.253	0.221	0.174	+15.618	20.930	21.112	0.351	0.294	+12.004
K_2O	0.180	0.181	0.002	0.001	0.520	0.529	0.006	0.004	+0.352	0.380	0.383	0.004	0.003	-0.102
CaO	0.330	0.331	0.006	0.003	0.820	0.833	0.015	0.012	+0.893	0.460	0.464	0.008	0.007	−0.478
Fe_2O_3	0.800	0.803	0.005	0.002	1.800	1.829	0.011	0.009	+0.666	1.970	1.987	0.012	0.010	+0.138
TiO_2	0.190	0.191	0.002	0.001	0.610	0.620	0.008	0.006	+0.499	0.340	0.343	0.004	0.004	−0.252
Cl	47.960	48.167	1.359	0.636	2.000	2.033	0.057	0.045	−59.126	0.220	0.222	0.006	0.005	−3.990
Σ	99.570	100.000	2.135	1.000	98.390	100.000	1.270	1.000		99.140	100.000	1.196	1.000	

Mole numbers of each compound (3) and their corresponding molar fractions (4). Columns 5a and 5b show respectively the values multiplied by 100 of the molar fraction variations between NMP and dross and between the crust and NMP. The positive sign indicates accumulation and the negative one, release.

generating droplets that try to pass through this salt and oxides viscous layer.

Another significant chemical component present in NMP and crust composition is the aluminum hydroxide (bayerite). This phase or α-$Al(OH)_3$ only takes place in the waste and crust composition due to the reactions already described in Eqs. (1) to (3) and to the hydrolysis of aluminum oxide (Eq. (5)) (Shinzato, 1999).

$$Al_2O_3 + 3H_2O \leftrightarrow 2Al(OH)_3 \quad (5)$$

The bayerite has a crystal structure in which the aluminum is octahedrally coordinated with double layers of hexagonal closed-packed hydroxyl groups stacked in an AB AB layer sequence (in gibbsite this sequence is AB BA). As other trihydroxides (gibbsite, nordstrandite) and oxyhydroxides (boehmite, diaspore), the bayerite can be used as a source of transition aluminas (TAs) (Santos et al., 2000). The family of transition aluminas is made up of all aluminas obtained by thermal decomposition of aluminum hydroxides or oxyhydroxides, with the exception of alpha-alumina. Those dehydration processes are irreversible, resulting in α-Al_2O_3 as an end product. Each hydroxide structure undergoes through at least two transition structures as the temperature rises before reaching the α-alumina structure. The thermal transformation sequence of bayerite is the following (Pechiney, 2003):

$$\alpha\text{-Aluminum hydroxide (bayerite)} \xrightarrow{280\ ^\circ C} \eta\text{-alumina (TA)} \xrightarrow{830\ ^\circ C} \theta\text{-alumina (TA)} \xrightarrow{1000\ ^\circ C} \alpha\text{-alumina(corundum)}$$

The transition aluminas are characterized by their micro-porosity and specific surface area. Therefore, TAs are commonly used for production of catalyst carriers and adsorbents.

5.2. Concrete blocks with NMP

The reduction in the working time of the blocks is probably a consequence of temperature increase resulting from the exothermic process caused by the cement hydration in the presence of particular constituents of NMP, such as alumina and metallic aluminum remainings. Mixtures of aluminous cement with Portland cement also set more rapidly, resulting in calcium sulpho-aluminate hydrate and calcium aluminate hydrate (George, 1983).

The flat texture of the blocks is probably a consequence of the size (<20 mesh) and roundness of the grains of NMP oxides. The rounded shape of the oxides is a result of the friction to which these materials were submitted in rotary drums during the leaching process.

The humidity test reveals that blocks are not hygroscopic, but the average value obtained in the absorption test for the studied blocks (11.15%) is slightly above that established by the specific standard (10%). All of the individual values of this test are in accordance with the norm (<15%).

Considering that NMP mainly consists of alumina, spinel and silica, the compressive strength values obtained for the tested blocks are probably the consequence of the low proportion of cement/aggregate used in their manufacture. The proportion usually recommended by the regulations is of 1:3 instead of 1:6 (used in the experiment) and, in this case, the proportion of cement decreased in 50%.

Despite the presence of some alkalis in NMP (Na_2O and K_2O) they can easily be eliminated by water leaching; the decrease of their concentrations in the crust (Table 4) confirms this fact.

The tests accomplished with this study are preliminary; additional tests are needed to fully evaluate the long-term stability and durability of these materials and also the aggregate/cement-paste interactions. These interactions produce the alkali-aggregate reactions (ARR) that occur with reactive aggregates (flints, opals, cherts, strained quartz, silicates and carbonates) and the alkali constituents (Na_2O and K_2O) of Portland cement, aggregates, external agents, etc. This reaction is driven by the high pH (>13) of the pore fluid that permeates the concrete microstructure giving rise to a gel product, causing swelling and expansion cracks in affected concretes (Barnett and Brew, 2004).

Although aluminas are not regarded as reactive aggregates, it is believed that they also react with alkaline solutions of concrete, due to the amphoteric behavior of aluminum. This fact is also confirmed by George (1983) that states that alumina trihydrate from aluminous cement becomes soluble in alkaline environments. It should be pointed out that alkali-aggregate reactions occur in ordinary concrete and also take place as from a certain number of years.

6. Conclusion

The leaching process used by tertiary aluminum industries produces a significant quantity of non-metallic wastes, which could be recovered if some improvements were included in this process. An efficient method of material washing, for example, could reduce the undesirable elements such as chlorides and alkalis.

The primary oxides of interest economically that are present in NMP are corundum, spinel and quartz. These constituents are quite resistant and corrosion inert, as well as very stable at high temperatures.

Another interesting chemical component present in these materials is bayerite, produced from the hydration of aluminum and its oxides. This chemical component can be used in the production of transition aluminas

that are commonly used as catalyst carriers and adsorbents.

The use of NMP as an additive or substitute of one portion of the aggregates in the manufacture of concrete blocks will be further evaluated in future studies, since preliminary results have shown low compression strength. Should a higher proportion of cement be employed, a better result could have been obtained.

However, a positive consideration based on this study refers to the other tests (dimensional, humidity and adsorption) and the acceleration of working time. This decrease in the working time will certainly result in an increase in the production of the concrete blocks, since they will be available for sale faster than the ones currently produced.

Another important use of NMP would be as a synthetic aluminous aggregate for refractory concrete, as well as cements of high alumina content specifically designed for high-temperature applications.

The commercial use of NMP can significantly reduce the quantity of waste disposed and also contribute to preservation of the environment. More over, using aluminum waste in preparing aluminum products, such as aluminum hydroxide, will certainly be more economically feasible than the use of bauxite.

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Mechanical properties of dewatered sewage sludge

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Abstract

The mechanical properties of dewatered, anaerobically digested sewage sludge were determined from soil laboratory tests. The sludge material is largely composed of organic clay sized-particles, a sizable fraction of which is in an active state of biological digestion which can continue over many years under field conditions. Moderately digested sludge material was found to have a typical specific gravity of solids value of 1.55, and loss on ignition (LOI) value of 70% dry mass. Strongly digested sludge, produced by digesting the liquid sludge further at 35 °C in the laboratory, was found to have a lower LOI value of 55% dry mass, and a higher specific gravity of solids value of about 1.72. The maximum dry density of 0.56 tonne/m^3 for the dried sludge material was produced using standard Proctor compaction at roughly 85% moisture content (54% solids content).

Air-dried, compacted sludge material was tested in quick-undrained triaxial compression and vane shear. Undrained shear strength–moisture content plots are presented. Shear strength values measured in triaxial compression and vane shear were consistent. The effective angle of shearing resistance (ϕ') was determined from consolidated-undrained, triaxial compression tests on pasteurized, normally consolidated samples of the sludge material. The mechanical properties of the sludge material changed with the level of sludge digestion. The ϕ' value increased from 32° for moderately digested sludge, to 37° for strongly digested sludge. The effective cohesion of the sludge material remained zero throughout.

The shrinkage, swelling and adhesion properties of the sludge material were also studied. Significant shrinkage occurred as the compacted material dried. The sludge material lost its adhesion below about 95% moisture content (51% solids content). Re-hydration of the dry material caused the bulk volume to double.
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1. Introduction

Sewage sludge is the slurry byproduct of wastewater treatment processes. More stringent controls on the quality of wastewater discharges are giving rise to ever increasing volumes of sewage sludge which must be safely disposed. One of the principal means of disposal is to landfill. The sludge material is usually mixing with other solid waste in municipal landfills. Sometimes the sludge material is disposed alone at dedicated monofill sites. For example, Russell (1992) reported one such monofill which was 25 m in height. The sludge material must be adequately dewatered for efficient disposal and to provide an adequate factor of safety against instability of the landfill slopes. The effects of ongoing biological digestion on the shear strength properties of the sludge material must also be considered when determining the long-term factor of safety against slope instability.

Current knowledge of the mechanical properties, in particular the shear strength properties, of dewatered sewage sludge is limited. In water treatment literature, the shear strength of the sludge material is often estimated from its solids content (SC) value, defined as the ratio of the dry mass of the solid particles to the bulk sludge mass, given as a percentage. In practice, however, shear strength estimates based on the solids content

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alone are unreliable. Casey (1991), for example, reported that depending on the source of the sludge material and the level of biological treatment, the solids content required for disposal of the sludge material in municipal landfills can vary between 15% and 50%.

Undrained shear strength–moisture content data were reported by Geuzens and Dieltjens (1991) for dewatered sewage sludge. Moisture content (w), used in geotechnical literature, is defined as the ratio of the mass of water in the void spaces to the mass of solid particles, given as a percentage. Such data are more useful in determining the actual level of dewatering required prior to landfilling the sludge material. Moisture content and solids content values can be related as follows:

$$\mathrm{SC} = \frac{100}{1 + \left(\frac{w}{100}\right)} \ [\%]. \tag{1}$$

Index, standard Proctor compaction and shear strength tests were conducted on samples of sewage sludge of various consistencies using standard soil laboratory test methods (Head, 1992, 1994, 1998). Biologically active samples were tested at different stages during the digestion process to study the effects of the initial level of treatment at the wastewater plant and ongoing digestion in the landfill on the mechanical properties. Samples of dewatered sewage sludge were obtained from Tullamore Municipal Wastewater Treatment Plant (Tullamore, Ireland) which serves an urban population of approximately 12,000. There was no significant industrial wastewater influent to the plant. The sewage sludge was biologically digested at the treatment plant using the anaerobic, activated sludge method (Metcalf and Eddy, 1991). The moisture content of the sludge material was reduced to roughly 720% (12% solids content) using belt process dewatering with polyelectrolytes added to the liquid sludge to encourage flocculation of the solid particles.

2. Index properties

The Atterberg limits, particle size distribution, specific gravity of solids, loss on ignition and pH of the sludge material were measured. Material direct from the wastewater treatment plant had a liquid limit of 315%, a plastic limit of 55%, and a plasticity index of 260%. When wet sieved, roughly 90% dry mass of the sludge material passed the 425 μm sieve indicating that the sludge largely comprised clay-sized particles. Material retained on this sieve comprised some organic fibers, grit particles and shredded plastic. The very high liquid limit and plastic limit values and colloidal activity (the plasticity index to clay fraction ratio) of the sludge are characteristic of calcium montmorillonite clay minerals (Yong and Warkentin, 1975). Similar liquid limit and plastic limit values were also reported for water treatment plant sludge by Wang et al. (1992) and for paper mill sludge by Charlie (1977). Ignition of dry, powdered sludge material at 440 °C caused a 70% reduction of the sample dry mass. This loss on ignition (LOI) value is an indirect but fairly accurate measure of the organic content of the sludge material. Based on the Atterberg limits and this LOI value, the sludge material is organic clay of high plasticity according to the Unified Soil Classification System.

The specific gravity of solids value of 1.55 was measured using the small pycnometer method. Kerosene was used instead of distilled water as the fluid in the pycnometers. Otherwise the sludge samples would have continued to slowly decompose during the tests yielding inaccurate results. The specific gravity of solids value of 1.55 is low when compared with inert soils which generally have a value in the range of 2.5–2.7. However, the specific gravity of solids value measured for the sludge material is consistent with the range of 1.4–2.1 reported for other sewage sludge by Campbell et al. (1978) and the range of 1.41–1.83 reported by Skempton and Petley (1970) for peat with similar LOI values. The pH value of the leachate was 8.0, which is slightly high for anaerobically digested sewage sludge. These LOI and pH values for the sludge material indicate that only moderate biological digestion of the sewage sludge occurred at the wastewater treatment plant. Strong digestion would have typically yielded an LOI value in the range 45–55% and a pH value in the range 6.6–7.4.

Index tests were also conducted on liquid sludge which was stored outdoors in drums and allowed to digest naturally. Two and a half years after the initial treatment at the wastewater plant, the specific gravity of solids value of the sludge material had increased from 1.55 to 1.63 while the LOI value had reduced significantly from 70% to 58%. The latter LOI value indicates that some further biological digestion of the sludge would continue to occur.

3. Compaction properties

Standard Proctor compaction tests were conducted on air-dried sludge material. The maximum dry density of 0.56 tonne/m^3 was achieved at roughly 85% moisture content (54% solids content), the optimum moisture content (OMC) for compaction. The bulk density and dry density values of the compacted sludge material were low but are consistent with the low range of specific gravity of solids values measured. The bulk density increased from about 0.65 tonne/m^3 for dry sludge material, which compacted poorly, to 1.10 tonne/m^3 for sludge material with moisture content values above OMC. More strongly digested sludge material yielded slightly higher bulk density and dry density values which

was due to the increase in the specific gravity of solids value with increasing levels of biological digestion.

The shrinkage, swelling and adhesion properties of the sludge material were also studied. The linear shrinkage value of 33% was measured as the percentage reduction in the length of a bar of the sludge material after it had been oven dried at 105 °C from the liquid limit condition. The sludge bar was rectangular in cross-section and initially 140 mm long. The swelling potential of the dry sludge material, which is sometimes produced in pellet form after thermal drying processes, was measured as the percentage increase in volume of the dry powered sludge material after it had been pluviated through water (Holtz and Gibbs, 1956). These free swell tests indicated that dry sludge material has the potential to approximately double in volume when fully rehydrated.

The lowest moisture content at which moist sludge material adheres to metal tools, often referred to as the sticky limit, was measured at roughly 95% moisture content (51% solids content). Sludge handling may prove difficult when the moisture content of the sludge exceeds the sticky limit value since the sludge material will tend to cling to the machine plant. The sticky limit value was measured as the lowest moisture content at which sludge particles adhered to a clean dry spatula, when the spatula was lightly drawn over the surface of a pat of the sludge material.

4. Shear strength properties

The short-term and long-term shear strength properties of the sludge material were measured in the laboratory. The short-term shear strength, usually referred to as the undrained shear strength, is pertinent to engineering the efficient handling of the sludge material, trafficability at the landfill site and an adequate factor of safety against slope instability during landfill operations. The effective-stress strength properties of the sludge material are used to calculate the factor of safety against instability of the landfill slopes for the intermediate and long-term conditions. Moderately and strongly digested sludge was tested to study the effects of biological digestion on the shear strength properties.

4.1. Undrained shear strength properties

The undrained shear strength of the dewatered sludge material was measured using the laboratory vane shear apparatus, with a 12.7-mm long by 12.7 mm wide, cruciform vane. The undrained shear strength of the dried, compacted sludge material was measured using both the triaxial compression apparatus and the vane shear apparatus. The sludge material was air-dried in the laboratory to moisture content values above the 85% OMC value and compacted into a 1-l mould using standard Proctor compactive effort. A set of three 38 mm diameter, triaxial samples were prepared from each of the Proctor samples. Some of the triaxial samples were air dried to reduce their moisture content below the OMC value before the samples were sheared. This sample preparation method yielded samples whose properties were characteristic of dried sludge material, compacted in the landfill under optimum conditions and allowed to air-dry. Fig. 1 shows the bulk density and dry density values of the samples tested in quick-undrained triaxial compression and vane shear. The compacted sludge material was partially saturated with an air voids content of about 5%.

An isotropic confining pressure of 100 kPa was applied to the triaxial samples which were then sheared quickly under undrained conditions at an axial compression rate of 1.6% strain/min. The shear stress–axial strain plots shown in Fig. 2 are characteristic of

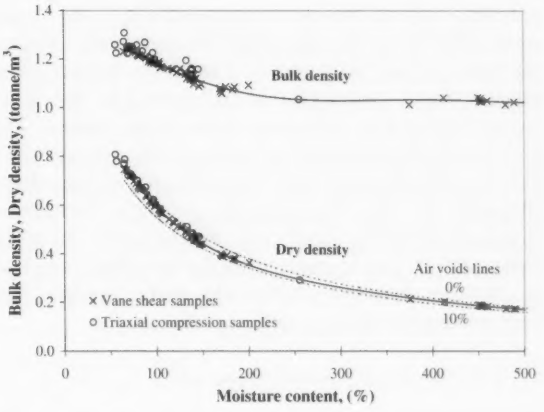

Fig. 1. Density vs. moisture content data for undrained shear strength samples.

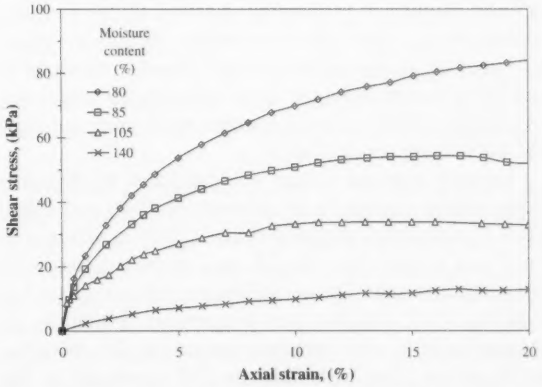

Fig. 2. Shear stress vs. axial strain data from quick-undrained triaxial compression tests.

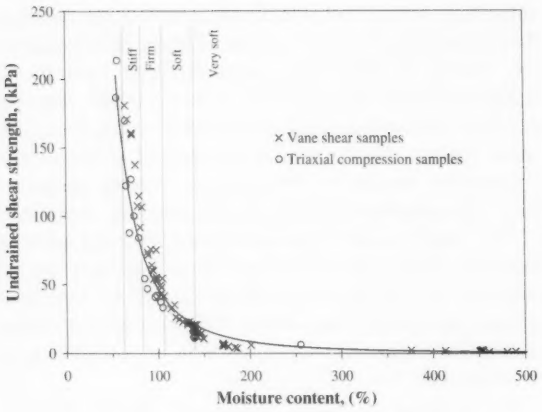

Fig. 3. Undrained shear strength vs. moisture content.

normally consolidated clay and indicate that large strains are required to fully mobilize the shear strength of the sludge material. The undrained shear strength–moisture content data summarized in Fig. 3 shows the usual behavior of the undrained shear strength increasing exponentially with reducing moisture content. The shear strength values plotted correspond to the shear stress mobilized at 20% axial strain of the sample. The undrained shear strength measured in triaxial compression was slightly lower than that measured in vane shear although the two data sets are generally consistent. Hence, in practice, quick and reliable shear strength measurements can be made using the vane shear apparatus in order to assess whether the sludge material has been adequately dewatered for landfilling.

4.2. Effective-stress strength properties

The effective-stress strength properties of moderately and strongly digested sludge material were determined using consolidated-undrained, triaxial compression tests and with measurement of the pore water pressure response. Samples of moderately digested sludge were obtained direct from the wastewater treatment plant. Cakes of the sludge material were formed by consolidating liquid sludge using the large hydraulic consolidation cell (Head, 1998). Triaxial samples were prepared from the pasteurized sludge cakes.

Strongly digested sludge was produced by digesting liquid sludge further in the laboratory. Two and a half liters of moderately digested sludge (720% moisture content) was poured into the 250-mm diameter, hydraulic consolidation cell. The consolidation cell was placed in a drying oven and the oven temperature gradually increased to 35 °C, the optimum temperature for mesophilic digestion. The consolidation cell remained in the oven for 21 days with the oven temperature held at 35 °C. Biogas evolved continuously at a rate of about 0.3 l biogas per day per liter of liquid sludge. In total, 23.7 l of biogas evolved and the volume of the liquid sludge sample was approximately halved. The oven temperature was gradually reduced back to laboratory temperature after the 21 days had elapsed. Index tests later confirmed that the sludge material was now strongly digested after this heat treatment. The specific gravity of solids value had increased from 1.55 to 1.72 while the LOI value had reduced from 70% to 55%.

The liquid sludge remaining in the hydraulic consolidation cell was consolidated one-dimensionally under an applied cell pressure of 100 kPa in order to form a sludge cake. The sludge cake was then pasteurized so that the sludge material would remain saturated, permitting accurate measurements of the pore water pressure response during the triaxial compression tests. The consolidation cell was gradually reheated to 80 °C in the drying oven and the sludge cake, contained in the consolidation cell, was allowed to swell under undrained conditions against the applied confining pressure of 100 kPa. After three hours of heating at 80 °C, biogas had stopped evolving from the sludge cake, indicating that pasteurization was complete. The oven temperature was gradually reduced back to laboratory temperature and the consolidation cell dismantled.

Two sets of three triaxial samples were prepared from pasteurized cakes of strongly and moderately digested sludge material. The 38-mm diameter samples were set up in the triaxial apparatus to facilitate drainage via the radial boundary and one end of the sample, with continuous measurement of the pore water pressure response. Cell confining pressure and sample back pressure increments were applied until a back pressure of 240 kPa was reached. Both sets of samples were then isotropically consolidated until effective confining pressures of 30, 60 and 140 kPa were achieved at the end of the consolidation stage. Volumetric strain–time plots recorded for the set of moderately digested samples are shown in Fig. 4.

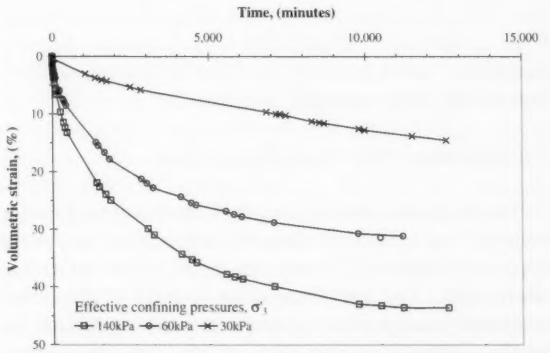

Fig. 4. Volumetric strain vs. time data from triaxial consolidation of moderately digested sludge.

The triaxial samples were sheared under undrained conditions at an axial compression rate of 0.002% strain/min which was sufficiently slow to allow all pore water pressures in the sample to equilibrate. Fig. 5 shows the effective stress path plots of $t = 1/2(\sigma_1 - \sigma_3)$ against $s' = 1/2(\sigma'_1 + \sigma'_3)$ for the set of three moderately digested samples. These stress paths are characteristic of normally consolidated clay. The test results were corrected for the effects of the sample membranes and side drains. The samples were deemed to have failed at 20% axial strain and the failure line of best fit was drawn through the origin and these set of points (Fig. 4). The value of the effective angle of shearing resistance (ϕ') for the moderately digested sludge of 32° was calculated using Eq. (2). Similarly, a ϕ' value of 37° was calculated for the strongly digested sludge material. This range of ϕ' values is similar to that reported for peat by Andersland et al. (1981). The effective cohesion remained zero, irrespective of the level of biological digestion.

$$\sin \phi' = \tan \alpha', \tag{2}$$

where α' is the angle of slope of the failure line to the horizontal s' axis (Fig. 4).

The ratio of the mobilized shear strength (c_u) to the effective consolidation pressure (σ'_c) was 1.1 at sample failure. This value equals that predicted for saturated normally consolidated clay using Eq. (3), after Skempton (1957). Similar c_u/σ'_c values were reported by Andersland et al. (1981) for model organic soils with comparable LOI values.

$$\frac{c_u}{\sigma'_c} = 0.11 + (0.0037)(\mathrm{PI}), \tag{3}$$

where PI is the plasticity index (%).

The effective angle of shearing resistance values of 32° and 37° were measured for pasteurized sludge material. However, these values can also be used to determine the long-term factor of safety against instability of landfill slopes constructed using biologically active sludge providing effective biogas control and leachate drainage systems are in place. Biogas which becomes trapped in the landfill body will cause a build up in the pore pressure, which if left unchecked, will cause swelling and potentially lead to slope instability (Russell, 1992). In other instances, further digestion of the landfilled sludge material can be hindered, for example, if industrial waste containing heavy metals were to become mixed with the sludge. Slope stability calculations should therefore be performed using the ϕ' values measured for both moderately and strongly digested sludge material. The long-term factor of safety will be given by the lower of the two factor of safety values calculated. The values of other mechanical properties used in these calculations must relate to the appropriate level of biological digestion.

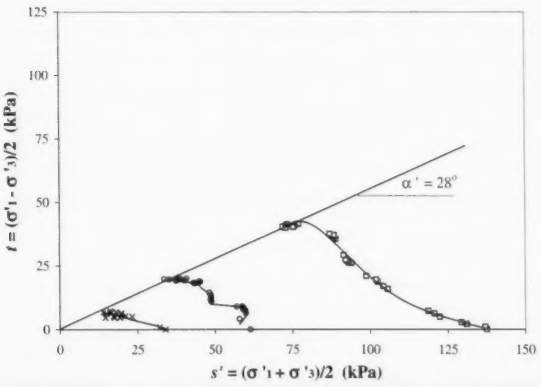

Fig. 5. Effective stress path plots for moderately digested sludge.

5. Summary and conclusions

The mechanical properties pertinent to the efficient handling and safe landfill disposal of dewatered sewage sludge were determined from soil laboratory tests. The material tested was an anaerobically digested sewage sludge which was found to be largely composed of organic clay sized-particles, a sizable fraction of which was still in an active state of biological digestion. However, the rate of digestion was highly dependent on the environment. For example, heating liquid sludge at 35 °C for 21 days under laboratory conditions produced similar levels of strong digestion as that achieved when the same material, stored outdoors in drums, was allowed to digest naturally at ambient temperature over two and a half years. Strongly digested sludge material was found to have a loss on ignition (LOI) value of about 55% dry mass, and a specific gravity of solids value of about 1.72. Moderately digested sludge material from the wastewater treatment plant had a lower specific gravity of solids value of 1.55 and a higher LOI value of 70% dry mass.

The maximum dry density of 0.56 tonne/m^3 for the sludge material was produced using standard Proctor compaction at roughly 85% moisture content (54% solids content). The density of the compacted material was low in comparison with inert soils but the density values are in line with the range of specific gravity of solids values measured for the sludge material.

Air-dried, compacted sludge material was tested in quick-undrained triaxial compression and vane shear. The undrained shear strength values measured in triaxial compression and vane shear were consistent. Hence, the vane shear apparatus can be used to quickly assess whether the sludge material has been dewatered sufficiently for landfilling. Undrained shear strength–moisture content plots are presented from which the level of dewatering necessary to facilitate efficient handling of the sludge material, trafficibility at the landfill site

and an adequate factor of safety against slope instability for the short-term condition can be determined. The effective-stress strength parameters were determined from consolidated-undrained, triaxial compression tests conducted on pasteurized, normally consolidated samples of the sludge material. The mechanical properties of the sludge material changed with the level of sludge digestion. The value of the effective angle of shearing resistance (ϕ') increased from 32° for moderate digestion, to 37° for strong digestion. The effective cohesion of the sludge material remained zero throughout. Providing effective biogas control and leachate drainage systems are in place, the factors of safety against instability of the landfill slopes for the intermediate and long-term conditions can be assessed using these ϕ' values.

The shrinkage, swelling and adhesion properties of the sludge material were also studied. Significant shrinkage occurred as the compacted sludge material dried. The sludge material lost its adhesion below about 95% moisture content (51% solids content). When the dry sludge material was full re-hydration, the bulk volume roughly doubled.

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Recycling rates of waste home appliances in Taiwan

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Abstract

The objective of this study is to provide a calculation of recycling rates of waste home appliances in Taiwan, for the EPA to amend these rates in order to increase the recycling efficiency. The recycling rate is calculated by a formula according to the statistical results obtained through: (1) an estimation of domestic use of home appliances using time series analysis with multiplicative seasonal ARIMA model, (2) a further estimation of generated waste home appliances based upon the estimated domestic use and the corresponding distribution of lifetime span, and (3) a cost analysis of recycling home appliances based on a sampling survey with stratified systematic sampling conducted among collectors and a survey on five recycling plants of waste home appliances. The suggested recycling rates in this study finally used by the EPA show that all of the recycling rates have increased compared with the rates in previous years. This study also implies that amending the recycling rates may only solve some recycling problems temporarily; however, the recycling system of waste home appliances in Taiwan has to be reformed to increase the recycling efficiency for the long term.
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1. Introduction

Recycling of solid wastes has increasingly become an urgent issue in Taiwan. The "Four-in-one recycling system" was implemented by the Environmental Protection Administration (EPA) of Taiwan in January 1997, to integrate four major players, namely, the people in our communities, the recycling management companies, the local governments, and the Recycling Management Fund (RMF) of the EPA, to increase the recycling efficiency. There are three organizations established under the recycling system, namely, the Fund Management Committee (FMC), the Fee Rate Review Committee (FRRC), and the independent auditing organization. In which, the FMC is organized by some committee members who are selected from enterprises' representatives, scholars, and experts. This committee is in charge of the operation of the RMF; while the FRRC consists of various representatives from different groups (e.g., environmental protection groups, consumer protection groups, scholars, experts, government authorities, enterprises, and EPA officials). The FRRC has the responsibility of considering the proclaimed recycling items [1] and the corresponding recycling rates. The missions of the FRRC contain the amendment, calculation, and adjustment for fee rates of recycling items. Moreover, the recycling plants should be audited and certified by those auditing agencies, and the RMF subsidizes those enterprises in accordance to the certified treated/recovered amount.

Under this recycling system, waste home appliances are required to be recycled. On July 5, 1997, the EPA

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[1] If the waste items are proclaimed to be recycled by the EPA, the corresponding responsible enterprises are enforced to pay recycling–clearance–disposal fees to the RMF for recycling the waste items.

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of Taiwan promulgated the recycling regulation of waste home appliances that include four items, namely, television, refrigerator, washing machine, and air conditioner. It is believed that this regulation pioneers the law enforcement of waste home appliances in the world. Under the current four-in-one recycling policy, manufacturers and importers of home appliances have to pay a recycling–clearance–disposal fee for each kind of the four items to the RMF based upon the rate approved by the EPA. Collectors and recycling plants receive subsidies from the RMF according to the collected and recycled (treated/recovered) amounts, respectively, audited and certified by an independent auditing organization. The subsidies are the major expenditure of the RMF.

In coordination with the recycling policy of the EPA, six recycling plants specialized in the treatment of waste home appliances following the guidelines of the EPA have been constructed during 1998–2001 (Chang et al., 2000), [2] which soon lead the recycling of waste home appliances in the world. Due to improper management of funding, the RMF had a serious financial imbalance problem in managing the waste home appliances at the beginning. Because of higher standard of regulation, the initial cost of these plants is much higher than that of other kinds of recycling business. Moreover, the market share of recycled materials is lacking and the final disposal of waste residues is costly. Recyclers can survive only due to the subsidy mechanism. About 75% of the operational income of the recycling plant comes from subsidy. It is argued whether or not the recycling plants are operated cost-effectively.

Nevertheless, the number of collected air conditioners, which contain polluted coolant, is much less compared with the other three items because of the higher recycled value. Most of the waste air conditioners have been sold at first place to the traditional dismantlers who are not appropriately equipped especially for the recovery CFCs and lubricants. The leaking of coolant has always been the major concern of environmental protection groups. On the other hand, the annual amount of collected waste home appliances is not growing after their storage at beginning phase have been treated/recovered; recycling plants start to concern the steady incoming of waste home appliances. In the mean time, collectors organized a collection center to store more waste home appliances and use it as a bargain to raise the selling price of waste home appliances to recycling plants, which was an impediment to the collection route.

[2] According to Chang et al. (2000), the EPA gave the guidelines for the construction of recycling plants through a series of workshops in 1998. Six plants operated by five companies were constructed following the guidelines.

All of these problems lead to some arguments on the recycling rates (or fees) of manufacturers/importers as well as the collection and recycling subsidies of collectors and recyclers, respectively, of these four waste items. How to amend a reasonable recycling rate becomes an important issue for the EPA. A proper recycling rate not only encourages recycling but also reaches the goal of waste reduction. This paper aims to provide a calculation of recycling rates to the RMF based on the principles of "polluters pay" and "economic efficiency". In order to calculate reasonable recycling rates, the collection and recycling costs have to be estimated based upon the sampling survey. Once the precise costs of collection/recycling are obtained, the recycling rates and subsidies could be set reasonably and be acceptable for both manufacturers/importers to pay and collectors/recyclers to receive. The results of statistical analysis provide helpful bases for amending recycling rates and subsidies of these four kinds of waste home appliances. This study provides four scenarios of recycling rates for each of the four waste home appliances based on two target recycling ratios and with or without taking the budget deficit into account. Finally, the official recycling rates of the EPA were amended based on the case of the target recycling ratio set at higher level, especially for air conditioners and taking the budget deficit into account. Comparing with the recycling rates in previous years, current rates were increased for all of four waste home appliances.

2. Recycling system of waste home appliances

The current recycling system of waste home appliances in Taiwan is based upon the "polluters pay principle (PPP)", which is different from other systems such as "extended producer responsibility (EPR)" and "integrated product policy (IPP)". The EPR system requires producers to have full obligation for recycling the products they produce. IPP seeks to improve the environmental performance by looking at all phases of the products' life cycle and taking action where it is most effective. However, with PPP, producers and importers in Taiwan have only obligation for paying the recycling–clearance–disposal fees to the RMF, but do not assume any responsibility for recycling, while the EPA undertakes the recycling work. The recycling system of waste home appliances in Taiwan is shown in Fig. 1. Waste home appliances discarded by consumers will be either recycled or disposed. The recycled waste home appliances were collected and transported to recycling plants or traditional dismantlers via various collection/recycling routes. Usually the collectors in the rear end of collection route are large-scale collectors who purchase waste home appliances from consumers, collection points, home appliance distributors, municipal solid

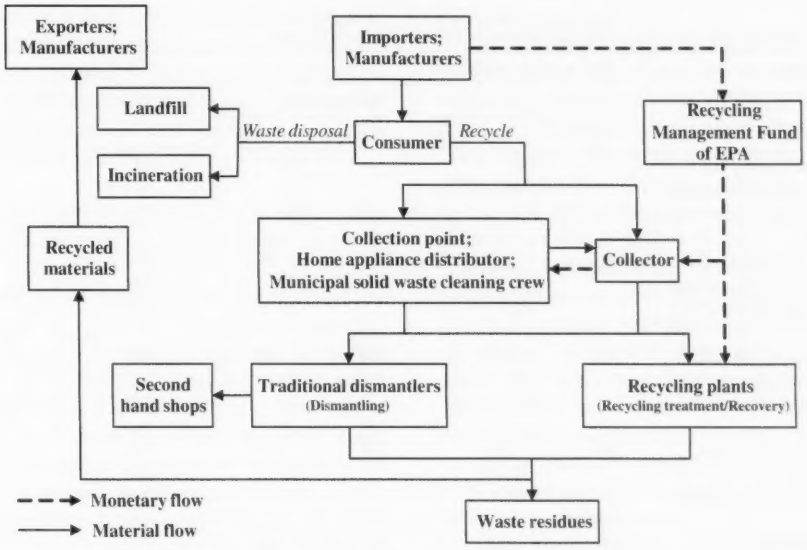

Fig. 1. Recycling system of waste home appliances in Taiwan.

waste cleaning crews, and the collectors in the front end of collection route with different prices. Recycling plants are required to have a permit [3] and special facilities especially for the waste refrigerators in order to receive recycling subsidy; whereas traditional dismantlers do not have a treatment permit.

Most of recycling plants have state-of-the-art treatment technologies, so that they can properly recycle hazardous substances such as coolants and PUR foaming agents in the waste refrigerators, coolant in the waste air conditioners, and fluorescent powder in the waste TV sets. Meanwhile the valuable recycled materials, such as iron, copper, aluminum, and plastic can be recovered and further reused. Traditional dismantlers without proper facilities can not suitably recover the hazardous substances. The valuable parts of waste home appliances such as compressors in air conditioners and refrigerators are separated and sold to second hand shops; whereas, improper treatment will leave problems of secondary pollution behind. Recycling plants could receive a recycling subsidy from the RMF for processing/recovering waste home appliances, but traditional dismantlers will not have any subsidy. Collectors will receive collection subsidy from the RMF if they ship the collected waste home appliances to recycling plants, but they do not get any subsidy for sending these goods to traditional dismantlers. Due to valuable parts of waste home appliances and a much lower treatment cost compared to recycling plants, traditional dismantlers may offer higher price to purchase waste home appliances and still be profitable without any subsidy from the EPA. If the price offered by traditional dismantlers is higher than the collection subsidy of the RMF, the collected waste home appliances will be sent to dismantling shops particularly waste air conditioners.

The recycling rates and subsidies implemented from March 1998 to December 2003 are shown in Table 1. The product-dependent charges on importers and manufacturers based upon the recycling rates approved by the FRRC and the amount produced/imported are the major sources of funds for the RMF. Moreover, the subsidies of collection paid to collectors are calculated from the certified amount multiplied by unit collection subsidy, while the subsidies of recycling paid to recycling plants are the certified amounts multiplied by unit recycling subsidy. The subsidies of collection and recycling are the major expenditures of the RMF. Basically, the recycling rates are calculated with a formula (will be discussed later) based upon the principle of 100% payment to those wastes generated in the domestic market; whereas the unit subsidies for collection and recycling are set by the EPA based upon the collection and recycling costs.

Since the official starting time for collectors to store waste home appliances was from June 1998, there were

[3] According to "The principle of subsidy for waste home appliances recycling" announced by the EPA on January 27, 2000, recycling plant only with "the Class-A plant permit" is qualified to apply for a recycling subsidy. The "collection–storage–clearance-treatment methods and facility standard of waste home appliances" announced by the EPA on December 25, 2002 took out the qualification limitation of "the Class-A plant permit" and replace the above subsidy principle. Therefore, before December 25, 2002, a Class-A treatment plant permit for waste home appliances from the EPA is required for receiving recycling subsidies. According to Chang et al. (2000), six plants have obtained the Class-A treatment plant permit.

Table 1
Yearly recycling rates and subsidies of waste home appliances in Taiwan (Unit: NT$/unit)

Year/month	Item	Waste home appliances					
		TV sets		Refrigerators		Washing machines	Air conditioners
		Big size	Small size	Big size	Small size		
1998/03–1999/06	Recycling rate		150		220	154	170
	Collection subsidy		180		393	244	275
	Recycling subsidy		330		410	220	145
1999/07–2000/04	Recycling rate	245	119	427	157	272	174
1999/07–2000/06	Collection subsidy		180		393	244	275
1999/07–2000/06	Recycling subsidy		330		410	220	145
2000/05–2001/08	Recycling rate	420	270	680	440	360	290
2000/07–2001/08	Collection subsidy		170		365	225	265
2000/07–2001/08	Recycling subsidy		330		410	220	145
2001/09–2001/12	Recycling rate	441	284	714	462	378	305
	Collection subsidy		170		365	225	315
	Recycling subsidy		280		370	190	120
2002/01–2002/12	Recycling rate	441	284	714	462	378	305
	Collection subsidy		127.5		302.5	175	302.5
	Recycling subsidy		252		333	171.5	108
2003/01–2003/12	Recycling rate	441	284	714	462	378	305
	Collection subsidy		127.5		302.5	175	302.5
	Recycling subsidy		252		333	171.5	108

Notes. (1) Collection subsidy includes collection fee, management fee of collection point, cleaning and transportation fee, management fee of storage, and storage fee from July 1998 to June 2000. (2) Collection subsidy includes collection fee, management fee of collection point, management fee of collector, transportation fee of collection, and transportation fee from collector to recycling plant for general area from July 2000 to October 2001. (3) Recycling subsidy includes process/recover subsidy, and only for recycling plant. (4) Recycling rates have taken deficit balance into account since September 2001. (5) The definitions of small size of TV and refrigerator are less than or equal to 25 in. (CRT size) and 250 l (volume), respectively.

no subsidies to the collectors before that time. Meanwhile, the subsidies for collectors were categorized into six items including collection fee, management fee of collection point, cleaning and transportation fee, management fee of storage, storage fee, and transportation fee from storage point to the recycling plant before September 2001. In which the storage fee was excluded from the subsidies and the subsidy for management of storage was renamed as management of collector from July 2000, because from then on the RMF stopped to support the storage of waste home appliances in responding to the operation of newly established recycling plants. To simplify the system of subsidy the five categories for collectors were combined into a collection subsidy from September 2001. Since the recycling plants officially started to operate from July 2000, the subsidies for recycling plants had never been used before that time; while the recycling rates approved by the FRRC in between March 1998 and June 1999 did not take recycling/treatment costs into account. In addition, the recycling rates set between July 1999 and April 2000 were too low to react to the real high recycling cost in the real operation, because of the underestimation of the recycling/treatment costs due to lack of field data. Therefore, after the recycling plants started to process, the accumulated stock of waste home appliances has been treated within one year. At that time, the recycling fund has come out a serious budget deficit because the income for recycling from manufacturers/importers could not afford the big amount of expenditure for recycling subsidies in a short time. The EPA tried to make up the problem of budget deficit by increasing recycling rates and reducing subsidies simultaneously. The recycling rates from September 2001 have taken deficit balance into account and subsidies have been reduced yearly from then on.

3. Methodology

The recycling rate is calculated by the following steps: (1) estimate the amount of waste home appliances; (2) estimate the collection and recycling costs based on the sampling survey; (3) calculate the recycling rate via a formula provided by the FRRC. The collection and recycling cost estimations of waste home appliances are mainly based upon the sampling survey, which includes collection routes, recycling/recovery treatment,

and annual amount of wastes generated; thereafter, statistical analyses are conducted by using these survey results. Estimation of amount of waste home appliances, sampling design of survey, economical/statistical analyses of collection/recycling costs, and the formula of recycling rate are explained in the following sub-sections.

3.1. Estimation for annual amount of domestic use, domestic generated waste, and recycled amount

The annual amount of domestic use is calculated by the annual amount of domestic production plus imported amount, minus the exported amount. Statistics on these data are not available yet during this research period (May–December 2001), therefore time series analysis was used to forecast the domestic use of home appliances in the year 2002. In response to the seasonal variation of consumption characteristic of home appliances, the multiplicative seasonal ARIMA model ARIMA$(p,d,q)\times(P,D,Q)_s$ (Vandaele, 1983) was used to fit the monthly data from January 1989 to December 2000.

Based upon the survey of the CTCI Foundation (1999), the relative frequency distribution of lifetime span of home appliances in Taiwan is shown in Table 2 (Hsu et al., 2001). The most frequent occurrence of lifetime span is in between 4 and 6 years for all home appliances except refrigerators, which is over 10 years. The domestic waste generated was estimated based on the estimated domestic use and their corresponding distribution of lifetime span.

The certified amount audited at the recycling plant for subsidy includes: (1) recycled amount collected by collectors and sent to recycling plants and (2) recovered amount of recycling plants. In this study, recycled ratio is defined as certified collected amount divided by waste generated amount; while recovered ratio is defined as certified recovered amount divided by waste generated amount. The recycled ratios in 2001 were 79%, 79%, 57%, and 16% for TV sets, refrigerators, washing machines, and air conditioners, respectively; while the recovered ratio in 2001 were, respectively, 83%, 84%, 55%, and 17%. Based upon the assumption that the recycling ratio in 2002 is the same as 2001, the estimated recycling amount in 2002 is derived from estimated waste generated in 2002 multiplied by recycling ratio in year 2001.

Table 2
Relative frequency distribution of lifetime span of home appliances in Taiwan (Unit: %)

Years	TV sets	Refrigerators	Washing machines	Air conditioners
1–3	6.51	5.32	8.35	8.61
4–6	36.87	26.58	36.53	57.23
7–9	20.44	28.58	29.45	20.28
10 and over	36.18	39.52	25.67	13.88
Total	100.00	100.00	100.00	100.00

3.2. Sample design

The aim of the present survey is to estimate the collection and recycling costs of waste home appliances in Taiwan. The base years of study are from 2000 to 2001. Target population includes all collectors and recycling plants of waste home appliances in Taiwan, which consists of 1141 collectors and 6 recycling plants. Stratified systematic sampling is used to select a probability sample of collectors; whereas, the corresponding estimator is used to estimate the collection cost. According to the fundamental principle of sampling survey theories (Cochran, 1997), if sampling units can be classified into strata that are internally homogeneous with respect to the variables to be estimated, and independent sub-samples are selected from each stratum, then the variance of overall estimators may be reduced. Due to the effect of socioeconomic characteristics and environmental policy of local government on the waste recycling system, it is commonly believed that the collection cost varies considerably with different counties. There are 23 counties in Taiwan; therefore, collectors were stratified by counties into 23 strata. A systematic sample was drawn within each stratum and selected collectors were interviewed to obtain related collection costs of waste home appliances. Six recycling plants already have permission to process/recover the waste home appliances, but only five plants were in operation during the survey period. Therefore, the five recycling plants in operation were investigated to obtain the recycling cost. Face to face interviews were conducted to collect data in order to increase the precision of estimation and reduce the non-response ratio.

3.3. Estimation of collection and recycling costs

The collection cost covers all the expenses that collectors collect waste home appliances from consumers and finally transport them to recycling plants. The major costs include annualized collection equipment cost as well as operation and management (O&M) cost (e.g., labor cost, storage cost, and administration cost). Equipment costs are one-time expenditures for vehicles and balers, which can be accounted as one-time expenses. Annualized equipment costs could present annual depreciation of equipment investment. Basically, equipment costs have been converted into annual costs by assuming a 7-year amortization period for collection equipment. Labor cost covers all the expenses of employee and employers. The storage cost is measured by rent of storage site for the collected materials. Administration cost includes haulage, equipment leasing and maintenance, licenses, insurance, as well as other administrative expenses.

The recycling cost includes intermediate process cost and the residue disposal cost. The intermediate process

cost covers all the expenses that recycling plants process/recover the collected waste home appliances. That represents recycling plants' actual intermediate processing expenses, which include annualized recycling capital cost, O&M cost (e.g., labor cost, equipment leasing and maintenance cost, rent, and administration cost). Capital costs are one-time expenditures including equipment (e.g., vehicles, storage containers, conveyors, crushers, balers, etc.), land, as well as building construction and improvements. Capital costs can be accounted as one-time expenses or amortized over the lifetime of the equipment. Annualized capital costs could present annual depreciation of capital investment. Basically, capital costs have been converted into annual costs by assuming 9-year and 16-year amortization periods for recycling/recovery equipment and building construction, respectively. Rent represents the expense for leased land and building, while the administration cost includes items such as transportation, licenses, insurance, loan interest, ISO, environmental safeguard, research, development, training, and other administrative expenses. Residue disposal cost represents the cost associated with final disposal of the waste. The administration cost does not take into account residue disposal cost. The residue disposal cost is calculated based on the contract between the recycling plant and the waste disposal agent. Most of the final waste residues are landfilled or incinerated; some small portion of final residues is temporarily stored and waiting for further process/treatment.

In order to increase the precision, the estimation of collection cost in this study considers about probability distribution, outliers, and scales of collectors simultaneously. The collectors in Taiwan have very different scales ranging from very small to very large. The collection cost may not be normally distributed. It is necessary to properly manipulate the outliers. Therefore, in order to increase the precision of estimation, a process was conducted to estimate collection cost for each stratum with the following steps:

1. Use Kolmogorov–Smirnov test (K–S test) to test the normality of the data.
2. If the data are normally distributed, then calculate the mean as an estimate of collection cost for that stratum.
3. If the data are not normally distributed, then conduct the following steps to obtain the estimate.
   (i) For data with symmetric or left-skewed distribution, use the method of α-Winsorized mean to adjust its distribution by choosing a proper α-value and obtain the mean for the adjusted data.
   (ii) For the data with right-skewed distribution, fit the data with Gamma or Lognormal distribution and obtain the mean of probability distribution.

Meanwhile, estimation formulas were used with the stratification of the sampling design to obtain the higher precision of estimation. The unbiased estimator of population mean is written as (Cochran, 1997; Hsu and Kuo, 2001)

$$\bar{y}_{st} = \left(\sum_{N=1}^{h} N_h \cdot \bar{y}_h\right) \Big/ N, \tag{1}$$

where $\bar{y}_{st}$ is the estimated mean of collection cost; N, the total number of collectors in population; N_h, the total number of collectors in stratum h; and $\bar{y}_h$ is the sample mean of collection cost in stratum h.

In Eq. (1), $\bar{y}_h$ was obtained through the estimation steps in previous descriptions. For stratified random sampling, the unbiased estimator of variance of estimated $\bar{y}_{st}$ is

$$\hat{V}(\bar{y}_{st}) = \sum_{N=1}^{h} \left[(N_h^2/N^2) \cdot ((N_h - n_h)/n_h) \cdot (s_h^2/n_h)\right], \tag{2}$$

where $\hat{V}(\bar{y}_{st})$ is the estimated variance of $\bar{y}_{st}$; n_h, the number of sampled collectors in stratum h; and s_h^2 is the sample variance of collection cost in stratum h.

The unit collection cost reflects annual cost of collection incurred in the base year of study divided by the annual amount of collected waste home appliances. The unit recycling cost reflects annual processing/recovering cost incurred in the base year of study divided by the annual amount processed/recovered. Recycling revenue is also calculated by the sale income of recycled materials. By subtracting recycling revenue from total recycling cost, the net cost of recycling is obtained.

3.4. Formula of recycling rate

The recycling fund obtained by law enforcement is used to support recycling activities, to encourage recycling of waste home appliances and to reduce the amount of municipal solid wastes. Under the four-in-one recycling system, the enforcement follows the "polluters pay principle", the manufactures/importers are mandated to pay recycling–clearance–disposal fees to the RMF based on the recycling rates approved by the FRRC and the amount that they produced or imported. The FRRC takes various factors, which include the recycling cost, reuse value, impact on environment, target recycling ratio, financial condition of the Fund, reward for recycling incentives, etc., into account to amend the recycling rates. In general, products for domestic use produced by domestic enterprises and imported by traders eventually will be either recycled or disposed. Thus, the recycling rates will be calculated based upon the principle of 100% payment to those wastes produced in the domestic market. The formula of recycling rates was developed by previous researches granted by the EPA, and was amended by the FRRC. In principle, the recycling rate is a function of this period's

collection and recycling costs, waste disposal cost, as well as last period's budget deficit

$$A = [B \times \alpha \times \beta + C \times (1 - \alpha) \times \beta] \pm D \times \beta, \quad (3)$$

where A is the recycling rate (NT$/unit); B, the recycling cost (NT$/unit) = (unit waste-purchasing cost for collectors (B_1) + unit collecting cost (B_2) + unit recovery cost (B_3) − unit value of recycled materials (B_4)); C, the weighted environmental cost (NT$/unit) = weighted cost of waste disposal cost, which is weighted by the ratio of incineration and landfill (C_1) + external environmental cost of waste (C_2); α, the target recycling ratio = annual recycled amount/annual waste amount; β, the waste generated ratio = annual waste amount/annual domestic sales amount = annual waste amount/annual declared business amount; D, the amortizing balance amount of recycling fund (NT$/unit) = reasonable balance amount of recycling fund/amortization period (year)/waste amount.

Basically, the recycling rate (A) includes two primary costs, namely, net recycling cost for those recycled waste home appliances (B) and weighted environmental cost for those as general wastes (C). In item B, collection cost, recovery cost, and prices of recycled materials would be influenced by the whole economy, and thus it reflects the reality of the recycling market. B is also the basis for setting collection and recycling subsidies. Therefore, this item should be adjusted and reviewed annually to comply with economic changes. Concerning potential environmental hazards associated with disposal, with this formula, the enterprises will not only pay the internal cost for recycling (B) and waste disposal cost (C_1), but also pay the external environmental cost (C_2) for those not recycled but disposed waste. The external environmental cost (C_2) represents the cost of pollution controls for incineration and landfill of solid waste, such as manipulation of wastewater, solid waste incineration ash, air pollution, etc. With original design, the target recycling ratio of waste home appliances (α) that should be recycled will be decided by the EPA based upon the real recycling situation and required level of environmental protection, which is, considering the following factors: toxicity of waste, pollution level, recycled value, the condition of recycled material market, and recycling technology. In order to reach the goal of efficient use of resource, the formula of recycling rate complies with the more difficult to be recycled and the less friendly to environment, the higher the rate is to be. Therefore, the target recycling ratios have usually been set up much higher than the estimated recycling ratios especially for those less environment friendly waste. In practice, if it is set too high to reach, the income of fund from product charges will be larger than the expenditure for recycling subsidy, which will result in a budget surplus. On the contrary, if it is set too low it will result in a budget deficit. Whatever, the recycling performance is usually criticized for both budget surplus and deficit. Therefore, the real recycling ratio is sometimes used instead of target recycling ratio to reach budget balance of the Fund.

In Eq. (3), the unit fee for balancing deficit (D) is used to balance the income and expenditure to avoid the deficit of the recycling fund. Concerning the budget balance, this item is included in the recycling rate formula. The amendment of fee rate should thus concern both the budget surplus and deficit. Especially, for those recycling items with a big amount of deficit, it is improper to amortize the remaining fund over only one year. This would put the enterprises under the risk of going out of business. Therefore, it is proper to consider a suitable amortization period for each kind of recycling item. In order to reach the goal of efficient use of resources, the formula for recycling rate complies with the more difficult to be recycled and the less friendly to environment, the higher the rate should be.

4. Results

4.1. Estimated annual amount of domestic use, domestic generated waste, and recycled amount

The estimated multiplicative seasonal ARIMA models of domestic use for waste home appliances are listed as follows:

$$\text{TV sets}: \text{ARIMA}(0,1,1)\times(0,1,1)_{12}$$
$$(1+0.28499B)\nabla\nabla_{12}Z_t = (1-0.75435B)(1-0.99999B^{12})a_t, \quad (4)$$

$$\text{Refrigerators}: \text{ARIMA}(0,1,1)\times(0,1,1)_{12}$$
$$\nabla\nabla_{12}Z_t = (1-0.73478B)(1-0.99998B^{12})a_t, \quad (5)$$

$$\text{Washing machines}: \text{ARIMA}(0,1,1)\times(0,1,1)_{12}$$
$$\nabla\nabla_{12}Z_t = (1-0.64805B)(1-0.99999B^{12})a_t, \quad (6)$$

$$\text{Air conditioners}: \text{ARIMA}(1,1,1)\times(0,1,1)_{12}$$
$$(1-0.34378B)\nabla\nabla_{12}Z_t = (1-0.78523B)(1-0.99999B^{12})a_t. \quad (7)$$

The resulting P-values as shown in Table 3 provide evidence that the above models selected to fit the data are quite appropriate. [4] The estimated models were used to forecast domestic use of home appliances in 2002 as shown in Table 4. The estimated annual amounts of domestic use for TVs, refrigerators, washing machines, and air conditioners are 1195, 615, 541, and 1120 thou-

[4] All the corresponding P-values of Q statistics bigger than 0.10 indicate the adequacy of models; while the P-value for each parameter smaller than 0.05 confirms that the parameter is significant.

Table 3
Estimated multiplicative seasonal ARIMA model of domestic use

Item	Parameter estimates and corresponding *P*-values		χ^2 test *P*-values for *Q* statistics			
	Estimates	*P*-values	$K = 6$	$K = 12$	$K = 18$	$K = 24$
TV sets	$\hat{\phi} = -0.28499$	0.0014	0.3481	0.4583	0.7505	0.6483
	$\hat{\theta} = 0.75435$	<0.0001				
	$\hat{\Theta} = 0.99998$	<0.0001				
Refrigerators	$\hat{\theta} = 0.73479$	<0.0001	0.1062	0.1780	0.2958	0.1648
	$\hat{\Theta} = 0.99999$	<0.0001				
Washing machines	$\hat{\theta} = 0.64805$	<0.0001	0.1563	0.4559	0.5801	0.5891
	$\hat{\Theta} = 0.99999$	<0.0001				
Air conditioners	$\hat{\phi} = 0.34378$	<0.0001	0.2185	0.4048	0.4920	0.7529
	$\hat{\theta} = 0.78523$	<0.0001				
	$\hat{\Theta} = 0.99999$	0.0148				

Table 4
Estimated annual quantities of waste home appliances for year 2002 (Unit: 1000 units)

	TV sets	Refrigerators	Washing machines	Air conditioners	Total
Amount of domestic use	1195	615	541	1120	3471
Amount of domestic generated waste	1030	701	548	1156	3435
Annual recycled amount	811	554	314	181	1860

Note. The quantities of domestic waste generated exclude the quantities exported and reused in second hand market.

sand units, respectively, in 2002. The forecast figures show that the amount of home appliances sold domestically is expected to be slightly lower in 2002 when compared with that of the previous year except for air conditioners.

The data in Table 4 also show the estimated domestic generated waste, and the amount recycled. According to the lifetime span, the annual amounts of domestically generated waste of TV sets, refrigerators, washing machines, and air conditioners are estimated at about 1030, 701, 548, and 1156 thousand units, respectively, in 2002. The estimated amount of domestic waste home appliances generated is also predicted as slightly reduced in 2002 compared with that of the previous year. In addition, results show that the amount of waste generated is larger than the domestic use except TV sets. This reflects the reality of consumption characteristic of home appliances in Taiwan, that is, the trend of amount goes down for domestic use and goes up for waste generation. Based upon the recycled ratios in 2001, 79%, 79%, 57%, and 16% for TV sets, refrigerators, washing machine, and air conditioners, respectively, the estimated annual recycled amounts for these four home appliances in 2002 are 811, 554, 314, and 181 thousand units, respectively. Totally, around 1860 thousand units of waste home appliances are estimated to be recycled in 2002. This result shows that if the recycling ratio is not increased the recycled amount is going to be reduced. However, the annual treatment capacity of the five recycling plants in Taiwan is around 3672 thousand units. Obviously, the recycling plants will face a shortage of recycling amount of waste home appliances.

The collection subsidy of collectors depends on the certified amount at the recycling plant where waste home appliances are shipped to. The shortage of recycling amount could leave the bargain space for the collectors to ask for some extra benefits from the recycling plants besides the collection subsidy from the RMF. The behavior of collectors might cause serious impediments in the collection route. One of the strategies discussed for solving this problem is proclaiming other items of home appliances for recycling. It is argued that more proclaimed recycling items may bring more problems. Moreover, the recycled ratio of waste air conditioners is much lower than that of others, because most of waste air conditioners were sold to traditional dismantlers. [5] Due to the higher recycled values of air conditioners, the price offered by traditional dismantlers is usually higher than the collection subsidy of the RMF. A higher collection subsidy may provide an incentive to collectors to send their collected air conditioners to recycling plants. This implies that increasing

[5] The recycled amount measured by certified amount does not take the treatment amount of dismantlers into account.

Table 5
Collection cost per-collector and recycling cost per-plant in the year 2001 (Unit: 1000 NT$/collector or plant)

	Collectors		Recycling plants	
	Mean	Standard error	Mean	Standard deviation
Annualized capital cost				
Land, building construction and its improvements	–	–	4818.4	4034.76
Equipment	74.3	7.6	14,175.6	1028.59
Labor cost	390.8	25.7	34,665.0	11,788.55
Management cost				
Equipment lease and maintenance	–	–	15,253.1	4342.44
Rent for storage site or land	59.2	6.6	12,191.5	5060.72
Administrative expense	194.5	20.5	30,394.0	5556.63
Total	718.8	49.4	111,497.6	14,336.19

Notes. (1) Annualized capital cost cover only equipment for collectors. (2) The above administrative expense for recycling plants did not include waste residue disposal cost.

the collection subsidy of waste air conditioners might be a better way to increase the recycling ratio and to solve the shortage of quantities of waste at the recycling plants as well.

4.2. Estimated collection costs, recycling costs, and recycling revenues

Tables 5 and 6 show the statistical results of collection and recycling costs based on the data obtained from the sampling survey. Table 5 presents the average collection cost per collector and average recycling cost per recycling plant, while Table 6 shows the unit collection and recycling costs. The data in Table 5 also reflect the associated cost incurred for collectors and recycling plants. The collection cost of waste home appliances in the base years of study (2000–2001) includes annualized equipment cost, labor cost, storage cost measured by rent, and administration cost. According to the survey of 66 collectors, the average collector incurred approximately NT$718,800 [6] for collecting about 5498 units of waste home appliances in 2001. Over 50% of the collection cost incurred was due to labor expenses. This result indicates the labor-intensive characteristic of waste home appliances collection in Taiwan.

Basically, five recycling plants purchased key facilities and recycling technologies from Germany (mainly CRT glass separation machinery and refrigerator shredder), and improved locally to comply with the local characteristics of waste home appliances in Taiwan. The treatment processes are similar for those plants with relative high standard of pollution control as required by the EPA. The treatment process for each of the four waste home appliances is described as follows: (1) For waste TV sets: first, dismantle the CRT from the enclosure, classify the parts; second, separate the cone tube glass and panel glass of the CRT; third, remove the fluorescent power from the panel glass; and then, recover and classify all the valuable materials. (2) For waste refrigerators: first, recover CFCs and lubricants; second, remove the compressor; third, shred the waste refrigerator in an enclosed condition, recover the PUR foaming agent and CFCs contained in the PUR; and then recover and classify all the valuable materials. (3) For waste air conditioners: first, recover the coolant and lubricants; second, remove the compressor; and then recover and classify all the valuable materials. (4) For waste washing machines: first, remove the motor from the main body; second, dismantle all the individual operation devices and shred the plastic cover; and then recover and classify all the valuable materials.

The results from the survey also present an average per recycling plant incurred around NT$111,498 thousand dollars for recovering about 381,488 units of waste home appliances in 2001. A little over one-third of the recycling cost incurred was due to labor expense and around 27% on administrative expense. The relatively low standard deviation of annualized equipment cost as shown in Table 5 indicates no major differences among annualized equipment costs of recycling plants. Nevertheless, the capital investment for land, building construction and improvements, and labor costs vary widely with high standard deviation for recycling plants. This is not surprising because cost of land use and labor between the southern and the northern parts of Taiwan are very different due to the different pricing level between urban and rural areas. In addition, the different level of environmental safeguard and pollution control among recycling plants is another reason. Differentiated recycling subsidies in response to different levels of environmental protection and pollution control have been discussed all the time.

The average collection cost as shown in Table 6 is about NT$73 per waste TV set, 176 per waste refrigerator, 146 per waste washing machine, and 118 per waste air conditioner. The collection costs are varied via different collection routes. The cheapest collection cost is via the route that the waste home appliance comes from the consumer via the home appliance distributor and directly transported to the recycling plant without going through any collectors. In Taiwan, only about 2% of the waste home appliances are collected through this route at a cost of NT$57 (22% reduction) per waste TV set, 144 (18% reduction) per waste refrigerator, 127 (13% reduction) per waste washing machine, and

[6] 1 USD = NT$34.87 as of May 22, 2003.

Table 6
Estimated collection/recycling costs of waste home appliances for the year 2002 (Unit: NT$/unit)

	TV sets	Refrigerators	Washing machines	Air conditioners
Averaged purchasing price of waste for collectors (B_1) (standard error)	105.36 (2.64)	215.89 (8.83)	210.51 (3.78)	125.98 (6.13)
Averaged collection cost of collectors (B_2) (standard error)	72.56 (14.77)	175.83 (35.78)	145.59 (24.70)	118.30 (29.62)
Averaged recycling cost of recycling plants (B_3) (standard deviation)	320.14 (62.72)	497.43 (101.95)	331.82 (58.81)	378.90 (82.42)
Averaged revenue of recyclers from selling recycled materials (B_4) (standard deviation)	23.18 (10.85)	161.30 (18.00)	91.92 (6.31)	297.49 (82.42)
Averaged residue disposal cost (standard deviation)	81.83 (26.70)	64.44 (17.24)	58.79 (39.93)	18.41 (11.99)
Estimated incineration/landfill cost (C_1)	26.40	26.80	29.38	27.88
Estimated external environmental cost of waste (C_2)	1.27	1.29	1.41	1.34

Note. B_3 has already taken residue disposal cost into account.

93 (21% reduction) per waste air conditioner. The home appliance distributors are part time collectors of waste home appliances who are playing an important role in the recycling route. Currently, most of home appliance distributors send the take-back waste home appliances to the nearby collectors because the distributors do not have enough storage space for the waste home appliances. Therefore, collectors act as transportation points where waste home appliances are temporarily stored and transported to a recycling plant whenever the collection amounts fulfill the transportation requirement. According to the survey, about 77% of waste home appliances are collected through the collectors. In order to reflect reality, the higher estimated collection cost based upon the collection cost of collectors is used for calculating recycling rates in response to the primary recycling route in Taiwan. The statistical results imply that reducing collection cost through changing the recycling route in practice is an alternative way to increase the recycling efficiency because both recycling rates and subsidies can be eventually reduced.

The unit recycling cost in Table 6 includes intermediate process cost and the residue disposal cost, which covers all the expenses that recycling plants process/recover a collected waste home appliance on average. In accordance with the survey, five operating recycling plants incur about NT$320, 497, 332, and 379 per unit for TV sets, refrigerators, washing machines, and air conditioners, respectively, to recover waste home appliances in 2001. The waste residues include cone tube glass, panel glass, and fluorescent powder from the CRT of waste TV sets; CFCs, mineral oil, and PUR foaming agent from waste refrigerators; CFCs and mineral oil from air conditioners; electronic scrap from these four waste home appliances. The residue disposal costs of TV sets, refrigerators, washing machines, and air conditioners are NT$82, 64, 59, and 18 per unit, respectively. Revenue per unit waste treated represents the total revenue received by the recycling plants from the sales of recycled materials divided by the total units of recycled/recovered home appliances. The revenue of recycled material generally includes metals such as iron, copper, and aluminum, as well as plastic materials recovered from these four waste home appliances; coolant and compressors recovered from waste refrigerators and air conditioners; motors recovered from waste washing machines. The revenue of recycling waste TVs, refrigerators, washing machines, and air conditioners are NT$23, 161, 92, and 297 per unit, respectively. The revenue is much less than the costs from conducting recycling because of the lower quality of recycled materials compared with virgin materials and less demand of recycled materials. The prices of recycled materials are much lower than those of corresponding virgin materials.

The recycling plants with higher levels of recycling/recovery technology could gather less waste residue and get higher revenues. In addition to the level of recycling technology of the recycling plant, the revenue of recycled material also highly depends on the domestic economic condition and international trading conditions. The net cost obtained by subtracting recycling revenue from recycling cost provides the base of subsidy for recycling plants. The net cost of recycling waste TVs, refrigerators, washing machines, and air conditioners are NT$297, 336, 240, and 81 per unit, respectively. The net recycling cost implies that the operation of a recycling plant is less cost-effective. These results also reveal that recycling plants of waste home appliances can only survive under the subsidy of the EPA in Taiwan. Obviously, the lower the residue disposal cost is, the higher the revenue of recycled materials is. Enhancing recycling technology to increase the quality of recycled materials, in fact, increases the demand for recycled materials and hence reduces disposal requirements. This is the better way to reduce the net recycling cost. Improved market conditions for recyclables, resulting from stimulated demand for recycled goods, also are alternatives for lowering net recycling costs. Obviously, eliminating waste collection and recycling costs are the key

issues to drive the recycling system to be cost-effective. However, under current PPP system, it is hard to reach the cost-effectiveness of recycling.

The weighted environmental cost for those home appliances as general wastes (C) reflects the internal and external environmental cost of incineration/landfill for waste home appliances, which is estimated based upon the following equations

$$C_1 = CC \times WU \times (1 - \gamma) \tag{8}$$

$$C_2 = EC \times WU \times (1 - \gamma) \tag{9}$$

where CC and EC are the waste disposal cost and external environmental cost of waste disposal per kilogram, respectively; WU is the weight of waste home appliance per unit; while γ is the average percentage of reuse of resources for a waste home appliance (Hsu et al., 2001). Waste disposal cost is estimated by weighting the cost of incineration and landfill based on the ratio of incineration and landfill. [7] The estimated waste disposal cost (CC) is NT$3.69/kg (Chien et al., 2001). The waste disposal cost in weight is transformed into unit cost based upon the unit weight and resource reuse ratio of waste home appliances. Based upon the benefit transfer approach, the cost of pollution controls for landfill and incineration of garbage in California was transferred to obtain the external environmental cost (EC) in Taiwan (Tellus Institute, 1991; Bor et al., 1997). [8] The estimated weighted environmental costs including internal and external costs are NT$27.67, 28.09, 30.79, and 29.22 per unit for TV sets, refrigerators, washing machines, and air conditioners, respectively.

4.3. Calculated recycling rates

Statistical results in Tables 4 and 6 are used for computing recycling rates. Substituting data of $B_1 \sim B_4$ and $C_1 \sim C_2$ in Table 6 into Eq. (3), obtains data B and C in Table 7. Table 7 provides four kinds of recycling rates $A_1 \sim A_4$ in response to alternative target recycling ratio and whether or not to balance the deficit of the recycling fund. Two target recycling ratios, α_1 and α_2 are provided for target recycling ratios. α_1 is set based on the recovered ratios in 2001 and the ratio of air conditioners is increased to 50% for the perspective of increasing recycling efficiency of waste air conditioners. The parameter α_2 is set based on the recycled ratios in 2001. Therefore, the selected target recycling ratios are, respectively, 83%, 84%, 55%, and 50% for TV sets, refrigerators, washing machines, and air conditioners for α_1 whereas 79%, 79%, 57%, and 16%, respectively, for α_2. Except for washing machines, the recovered ratios are larger than the recycled ratios because some of the recovered waste home appliances are from storage collected in previous years. The four recycling rates provided in this study are: (1) the recycling rate (A_1) calculated based upon setting the target recycling ratio atα_1 without taking deficit balancing into account; (2) the recycling rate (A_2) calculated based upon setting the target recycling ratio at α_1 taking deficit balancing into account; (3) the recycling rate (A_3) calculated based upon setting the target recycling ratio at α_2 without taking deficit balancing into account; and (4) the recycling rate (A_4) calculated based upon the target recycling ratio at α_2 taking deficit balancing.

In order to release the pressure from expenses on storage, recycling plants were asked by the EPA to accelerate the recycling treatment using two shifts, which also caused serious deficit of the RMF. In order to balance the serious deficit of the fund, it is suitable to take a proper amortized amount of budget balance into account. Basically, the amortized amount of budget balance is taken based upon the principle that the percentage change of calculated recycling rate compared with the previous period is within 5% to avoid large impact on associated product market. The amortized amounts of budget balance into account are NT$−62, 79, −29 and −123 per unit for TV sets, refrigerators, washing machines, and air conditioners, respectively, with A_2 while NT$−113, 145, −53, and −225 for that of A_4. The calculated recycling rates presented in Table 7 have various differences among the four cases for each of the four home appliances, in which, A_2 is a better choice to increase the recycling efficiency and release the pressure from budget deficit as well. By choosing A_2 the recycling rates have to be increased for all four waste home appliances compared with the previous recycling rates for the period before September 2001. Policymakers may choose the suitable recycling rates based on their policy concerns.

Current recycling rates (from September 2001 to December 2003) as shown in Table 1 are under the condition of A_2 with some modifications for policy concerns. That is, current recycling rates of waste home appliance are calculated based upon higher target recycling ratio (α_1) and taking the budget deficits into account. Concerning the importers' and manufacturers' economic loading, the rates for small size TV sets and refrigerators are reduced. Since most of waste air conditioners have been sold to traditional dismantlers, the recycling rate for air conditioner is adjusted upwards to gather more money for the need of increasing subsidy to encourage the recycling of air conditioners. Finally, the current recycling rates for the big (small) TV sets, big (small) refrigerators, washing machines, and air

[7] Chien et al. (2001) conducted a cost survey to estimate the landfill and incineration costs in Taiwan.

[8] The external cost of landfill and incineration of garbage in California is from Tellus Institute (1991). Bor et al. (1997) estimated the external environmental cost for Taiwan by adopting the cost of pollution control of California and transferring it according to the currency exchange, price inflation index, GNP, and density of population to adapt to the Taiwan circumstance.

Table 7
Calculation of recycling rates of waste home appliances for the year 2002

	TV sets	Refrigerators	Washing machines	Air conditioners
Net recycling cost (B) (NT$/unit)	474.88	727.85	596	325.69
Cost of general wastes (C) (NT$/unit)	27.67	28.09	30.79	29.22
Target recycling ratio (α_1) (%)	83	84	55	50
Target recycling ratio (α_2) (%)	79	79	57	16
Discarded ratio (β) (%)	81	113	102	100
Amortized amount of budget balance for A_2 (NT$/unit)	−62	79	−29	−124
Amortized amount of budget balance for A_4 (NT$/unit)	−113	145	−53	−225
Recycling rates (A_1) (NT$/unit)				
Big size	370	805	349	177
Small size	247	537		
Recycling rates (A_2) (NT$/unit)				
Big size	441	714	378	301
Small size	294	476		
Recycling rates (A_3) (NT$/unit)				
Big size	353	758	361	76
Small size	235	505		
Recycling rates (A_4) (NT$/unit)				
Big size	483	591	414	301
Small size	322	394		

Notes. (1) A_1: Recycling rate of setting target recycling ratio at α_1 but not taking deficit balancing into account. (2) A_2: Recycling rate of setting target recycling ratio at α_1 and taking deficit balancing into account. (3) A_3: Recycling rate of setting target recycling ratio at α_2, but not taking deficit balancing into account. (4) A_4: Recycling rate of setting target recycling ratio at α_2 and taking deficit balancing into account.

conditioners are NT$441 (284), 714 (462), 378, and 305, respectively, which have been in effect since September 2001. Compared to the recycling rates in previous years, all the recycling rates have increased. It is expected that current recycling rates will more or less solve the recycling problems in previous years.

5. Conclusions and policy implications

The recycling system of waste home appliances in Taiwan has been operating for over five years. Under the four-in-one recycling system, current recycling rates used for supporting recycling activities of waste home appliances follow "polluters pay principle", and are calculated by a formula. In order to calculate recycling rates, this study investigates the collection and recycling costs of waste home appliances in accordance with these newly established routes and facilities, respectively. The calculated recycling rates with A_2 were suggested to increase the recycling efficiency and release the pressure from budget deficits. Finally, current recycling rates for TV sets, refrigerators, washing machines, and air conditioners, which have been in effect since September 2001 were set based on the suggested recycling rates in this study with some small changes concerning policies. Comparing with the recycling rates in previous years, all of the recycling rates have increased. In order to solve the environment problem associated with the leaking of coolant due to improper treatment by traditional dismantlers, the current recycling rate approved by the FRRC for air conditioners is adjusted upwards of NT$4 per unit to collect more money for the need to increase the subsidy to encourage the recycling of waste air conditioners. Increasing the collection subsidy of waste air conditioner is suggested to be a better way to increase the recycling ratio and solve the shortage of waste amount for recycling plants as well.

Statistical results in this study provide an insight into the cost components of recycling. Such insights may provide useful indications of how to improve future collection route and recycling process of waste home appliances to reach the goal of cost-effectiveness. Simplifying the current recycling route to reduce the collection cost as well as enhancing recycling technology to increase the quality of recycled material and to reduce recycling cost is suggested to be a better way to reach cost-effectiveness. Of course, the encouragement of using recycled material instead of virgin material is also an alternative option to increase recycling efficiency.

The results also provide some policy implications to the EPA for the future policy reform of recycling system of waste home appliances to achieve the goal of cost-effectiveness. However, the increased recycling rates may only temporarily solve the budget deficit of the fund. Under the current "polluters pay principle" system, it is hard to reach the cost-effectiveness of recycling. The management of product charge and subsidies of recycling can only solve the problem temporarily. In a short term, the "extended producer responsibility" system is suggested to be a better way to increase the recycling efficiency. However, in order to further reduce waste and increase the recycling efficiency, "integrated product policy" system might be a better way for the long term, for example, by stimulating the consumption demand on

products with green mark or with high percentage of recycled material by price mechanism. This may lead the producers to develop more environmentally friendly products and indirectly help in avoiding waste generation.

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Utilization of magnetic and electrostatic separation in the recycling of printed circuit boards scrap

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Abstract

The progress of the technology is directly related to the growth of production and consumption of electrical/electronics equipment, especially of personal computers. This type of equipment has a relatively short average lifetime, 2–3 years. The amount of defective or obsolete equipment has been increasing substantially; consequently its disposition and/or recycling should be studied.

In this work, printed circuit boards, which are used in personal computers, were studied in order to recover the metals in the circuit boards through mechanical processing, such as crushing, screening, as well as magnetic and electrostatic separation.

The results obtained demonstrate the feasibility of using these processes to separate metal fractions from polymers and ceramics, and that it is possible to obtain a fraction concentrated in metals containing more than 50% on average of copper, 24% of tin and 8% of lead.
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1. Introduction

The quantity of electro/electronic equipment produced nowadays is very high and it has been constantly increasing due to the growth in the consumption of these goods by the population as well as the relatively short lifetime of this equipment.

On the basis of its high electrical conductivity, copper is the one of the basic materials of electrical engineering technology.

Bertram et al. (2002) studied the waste management system, especially the copper cycle in Europe, and they affirm that wastes from electrical and electronic equipment (WEEE) are the fastest growing waste category; this finding emphasizes the need for efficient WEEE recycling strategies.

In western Europe, 6 million ton of electric and electronic waste were generated in 1998. The amount of this waste is expected to increase by at least 3–5% per annum (Forssberg and Cui, 2003). According to Bertram et al. (2002), the generation of WEEE is around 7 kg per capita per year in Europe. In the USA, a recent study predicted that over 315 million computers would be at the end of their life by the year 2004 (Forssberg and Cui, 2003).

In Taiwan, in terms of industries producing post consumer wastes, the top three are electronics and electrical engineering industries, with 630.000 ton (43%), chemicals with 450.000 ton (31%) and metal industries with 200.000 ton (14%) (Wei, 2000).

The recycling of this scrap is still quite limited due to the heterogeneity of the materials present on the product and the complexity of the production of this equipment.

Printed circuit boards are part of this equipment and their composition is quite varied, containing polymers,

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ceramics and metals. The metal content is around 28% (copper: 10–20%, lead: 1–5%, nickel: 1–3%). The content of the most important materials remaining is: plastics 19%, bromine (especially as flame-retardant) 4%, glass and ceramics 49% (Ludwig et al., 2003). In addition, there are also considerable quantities of toxicologically relevant substances, such as the flame-retarding "synergist" antimony trioxide, which is suspected to be carcinogenic, as well as precious metals (in decreasing order: silver, platinum metals like palladium, gold, to a total of ≈0.3–0.4%).

Aside from the special case of precious metals, copper in circuit boards is economically the most interesting metal for recovery and use as a secondary raw material because of its relatively high content compared to the corresponding content in explorable ores and its economic value. Copper recovery is also of ecological importance because of the relatively high copper content in comparison to the earth's crust composition and the corresponding low acceptable concentration limits, concerning leachate from landfills (Ludwig et al., 2003). Nevertheless, the contents of some chemical elements are much higher than in earth's crust, especially of some harmful chemical elements like bromine, antimony, cadmium and lead.

In addition to these inorganic elements, the following important organic compounds are found in circuit boards: isocyanates and phosgene from polyurethanes, acrylic and phenolic resins, epoxides and phenols, such as chip glues (Ludwig et al., 2003). Therefore, the circuit boards cannot be considered inert materials and are not suited for direct disposal in landfills.

The existing processes of recycling printed circuit boards use pyrometallurgical (Felix and Riet, 1994; Schichang et al., 1994; Bernardes et al., 1997; Szczygiel et al., 1998) or hydrometallurgical methods (Gloe et al., 1990; Pozzo et al., 1991; Hoffmann, 1992; Shibata and Matsumoto, 1999; Marca et al., 2002), which generate atmospheric pollution through the release of dioxins and furans (Menad et al., 1998) or high volumes of effluents.

Some authors are using mechanical processing (Zhang and Forssberg, 1997; Tenório et al., 1997; Noakes, 1999; Veit et al., 2002a) as an alternative to concentrate the metals in a fraction and the polymers and ceramics in another. The metal concentrate fraction can be sent to electrochemistry processes (Brown, 1992; Scott et al., 1997; Fornari and Abbruzzese, 1999; Kékesi et al., 2000; Brandon et al., 2002; Ubaldini et al., 2003) in order to separate the metals among themselves.

This work intends to study magnetic and electrostatic separation processes in order to obtain fractions with higher metals concentration, especially copper, from printed circuit boards scrap, previously comminuted and separated by size.

2. Experimental

In this work, mechanical processing was used as an alternative to processes that are usually used (thermal and hydrometallurgical processes) to the recycling of copper. This was done in order to concentrate metals in a fraction and polymeric and ceramic materials in another, without great amount of energy consumption and atmospheric pollution generation, case of thermal processes, or great amount of effluents generation, case of hydrometallurgical processes.

The steps of mechanical processing used in this work were comminution, size separation, magnetic separation and electrostatic separation.

All experiments were conducted at least two times and the analysis show an experimental error always smaller than 5%.

2.1. Preparation, comminution and size separation

Particle size, shape and liberation degree play crucial roles in mechanical recycling processes. Almost all mechanical recycling processes have a certain effective size range (Forssberg and Cui, 2003).

Printed circuit board scrap used in this work originated from damaged or obsolete personal computers. About 3 kg of whole printed circuit boards as well as 4 kg of electronic components were used. By previous experiences, we accepted these as a representative sample to this study, since there are no great differences in copper content between printed circuit boards in new and older personal computers (Angerer et al., 1993; Veit et al., 2002a,b). Each motherboard of personal computers weighs 500 g on average. With 3 kg of printed circuit boards, we are working with scrap of six personal computers. The average metal composition found in printed circuit boards was previously studied (Veit et al., 2002a). The copper content of this scrap is on average 12.5%. Besides this scrap contains 4.0% tin and 2.7% lead.

The electronic components used in this work were removed from the substrata, to which they were welded, by heating the printed circuit boards and consequently melting the Pb–Sn weld. This way we obtained two types of samples:

- Entire printed circuit boards (containing also electronic components) (PCB): ±3 kg.
- Electronic components (just the electronic components) (EC): ±4 kg.

These two types of samples were used in order to evaluate two different possibilities of obtaining the scrap. The first one occurs with post consumer (defective or obsolete) personal computers. The second sample could be generated with residues of the production line,

when defects are detected or some project changes are necessary.

Both types of samples were comminuted separately in a cutting mill until the fractions reached particle sizes smaller than 1 mm. The scrap was comminuted below 1 mm to warrant an excellent grade of release from metals (Zhang and Forssberg, 1997).

The samples were separated by particle size into three different fractions: F1 < 0.25 mm; 0.25 < F2 < 0.50 mm and 0.50 < F3 < 1.00 mm.

The chemical composition of the scrap studied is presented in Table 1 (for the entire printed circuit boards) and in Table 2 (for the electronic components). From the data in the tables, it is possible to say that copper is the main element present in printed circuit board scrap, reaching 24% in F3 of whole printed circuit board and 30% in F3 of electronic components.

Lead, tin and aluminum also have significant concentrations: 3.2% lead in electronic components, 4.9% tin in electronic components and 3.0% aluminum in whole printed circuit board.

2.2. Magnetic separation

When a magnetisable particle is placed in a magnetic field, it is acted upon by a magnetic force. Various classification schemes of magnetic separators have been introduced and probably the most practical and logical one is that which classifies separators as either dry or wet.

The equipment used was a dry magnetic separator, with a maximum magnetic field of 6500 G. A dry separator was chosen since for this type of residue the operation is easier; in addition the scrap was hydrophobic and would therefore, make the use of a wet separator more difficult.

A dry magnetic separation process was applied to each fraction of each sample, using, on average, a magnetic field from 6000 to 6500 G. A magnetic field close to the maximum capacity of the equipment was chosen to guarantee that all magnetic material was separated, since due to the small amount (in terms of mass) the magnetic material could be dragged with other non-magnetic materials present in the scrap.

Table 1
Chemical composition of complete printed circuit boards after comminution and particle size separation (Veit et al., 2002b)

Chemical composition (%)	F1	F2	F3
Copper	6.28	23.53	24.34
Iron	0.13	0.13	0.18
Aluminum	3.01	1.55	1.56
Nickel	0.05	0.20	0.20
Lead	0.35	0.95	1.35
Tin	2.51	2.50	2.51

F1 < 0.25mm; 0.25 < F2 < 0.50 mm; 0.50 < F3 < 1.00 mm.

Table 2
Chemical composition for electronic components after comminution and particle size separation (Veit et al., 2002b)

Chemical composition (%)	F1	F2	F3
Copper	9.68	17.25	30.15
Iron	0.28	0.10	0.80
Aluminum	2.06	1.62	0.92
Nickel	0.40	0.31	0.60
Lead	2.34	3.18	2.78
Tin	3.83	4.78	4.92

F1 < 0.25mm; 0.25 < F2 < 0.50 mm; 0.50 < F3 < 1.00 mm.

The main application of dry magnetic separators is either the removal of tramp iron and strongly magnetic impurities or the concentration of a strongly magnetic valuable component (Svoboda and Fujita, 2003).

In this stage, we obtained a magnetic fraction and a non-magnetic fraction. The non-magnetic fraction was transported to an electrostatic separator that would separate conducting from non-conducting materials.

2.3. Electrostatic separation

The electrostatic separators were initially developed to recover non-ferrous metals from shredded automobile scrap or for the treatment of municipal solid waste, but are now widely used for other purposes including foundry casting sand, polyester polyethylene terephthalate (PET), electronic scrap, glass cullet, shredder fluff, and spent potliner.

The electrostatic separator segregates conducting of non-conducting materials. The equipment includes two electrodes of continuous current, one of induction and another of attraction. The extreme difference in the electrical conductivity or specific electrical resistance between metals and non-metals provides an excellent condition for the successful implementation of a corona electrostatic separation in waste recycling. This separation has been mainly utilized for the recovery of copper or aluminum from chopped electric wires and cables, and the recovery of copper and precious metals from printed circuit board scrap.

The electrostatic separator used was the Model ES 1010 produced by Equimag. Fig. 1 shows schematically the operation of the electrostatic separator.

In this work, the electrostatic separator was controlled as follows:

- Ionizing electrode: distance of rotor = 25 cm and angle = 80°.
- Static electrode: distance of rotor = 25 cm and angle = 52.5°.
- Rotation of the rotor: 85 rpm.
- Source of high tension: 45–46 kV.

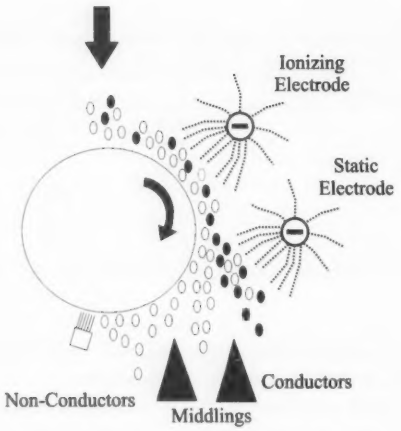

Fig. 1. Operation of an electrostatic separator.

The initial parameters were chosen according to studies by Zhang and Forssberg (1998). Some adjustments were done in order to achieve better results, what was observed by practical experiments.

A conductive fraction and a non-conductive one resulted from the electrostatic separation. The flow chart (Fig. 2) shows schematically all of the experimental steps of this work.

The magnetic, conductive and non-conductive samples were dissolved with aqua regia and after were chemically analyzed by means of atomic absorption spectrometry (AAS).

3. Results

3.1. Magnetic separation

The results are presented in Fig. 2, which shows quantities (mass) of magnetic material and non-magnetic material in each fraction after magnetic separation for whole printed circuit board and electronic components. Fig. 3 presents the contents of Fe, Ni, Cu and Pb after chemical analysis. The metals that should be analyzed were chosen as follows: in Fe

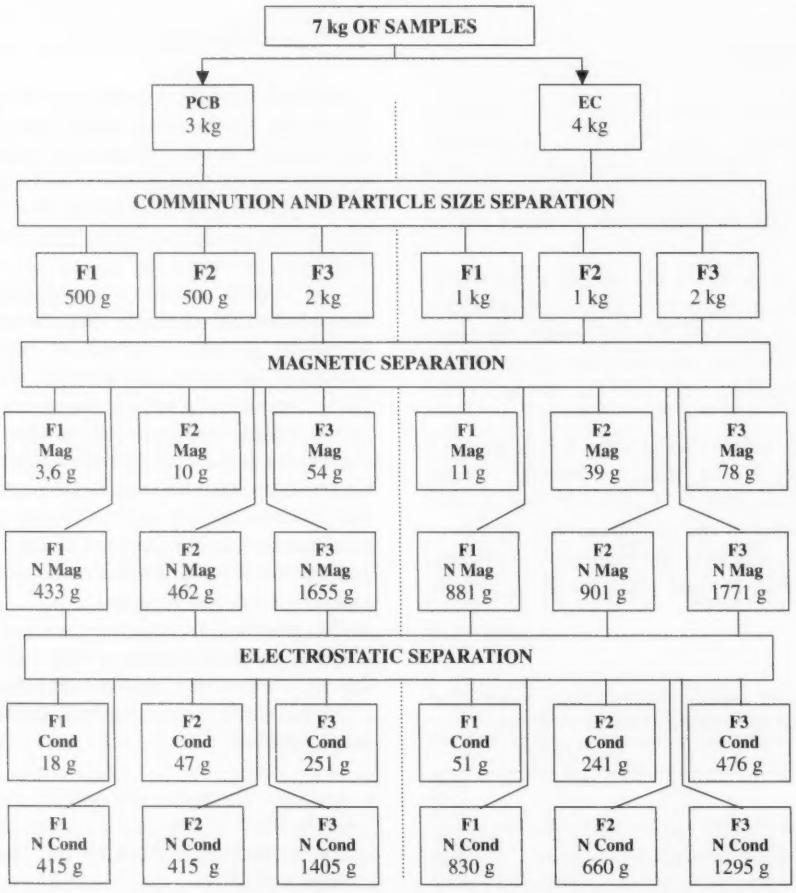

Fig. 2. Flowchart of the printed circuit board scraps recycling process. Mag, magnetic; N Mag, non-magnetic; Cond, conductive; N Cond, Non-conductive; PCB, printed circuit board; EC, electronic components; F1, $f < 0.25$ mm; F2, $0.25 < f < 0.50$ mm; F3, $0.50 < f < 1.0$ mm.

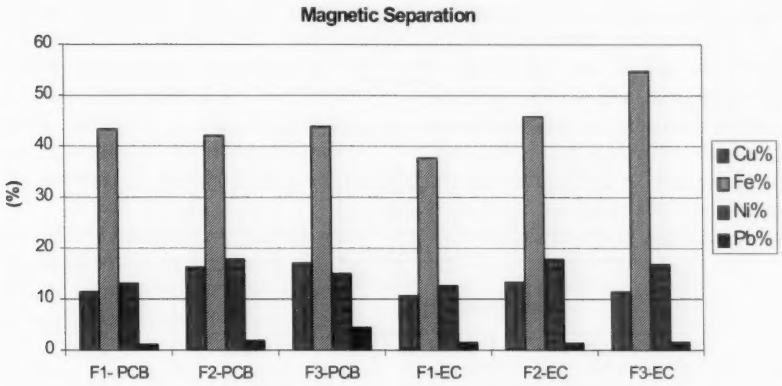

Fig. 3. Chemical analysis of the magnetic fractions after magnetic separation.

and Ni cases, because they are ferromagnetic, they should be present after a magnetic separation; in the case of Cu, since this is the most commonly present element in printed circuit boards and the main metal studied in this work; in the case of Pb, because it could turn the waste of our recycling process into a hazardous waste.

3.2. Electrostatic separation

Fig. 2 presents quantities (mass) of conductive and non-conductive materials in each fraction after an electrostatic separation for whole printed circuit board and electronic components. The following figures present the percentages of conductive materials in each

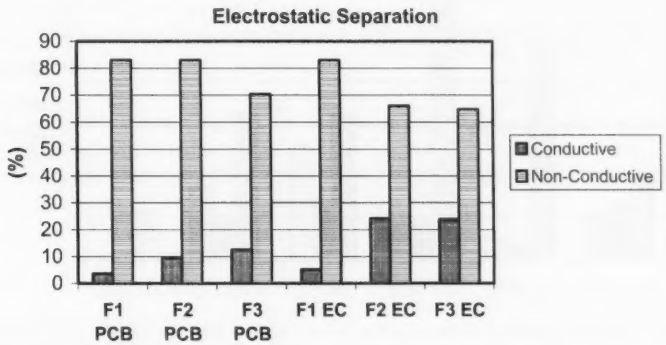

Fig. 4. Percentage of conductive and non-conductive materials after electrostatic separation.

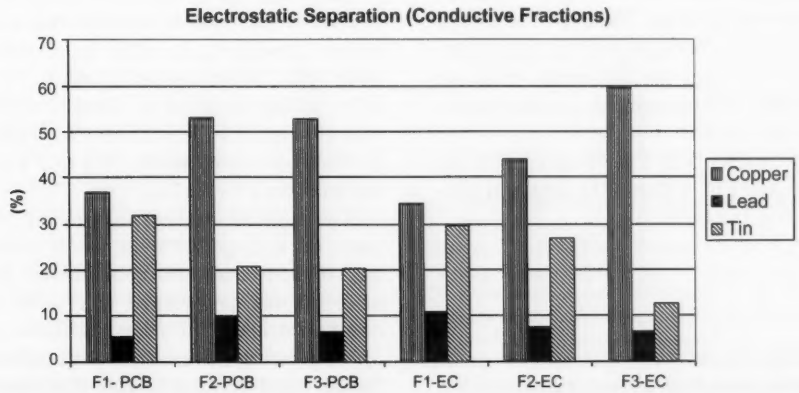

Fig. 5. Chemical analysis of the conductive fractions after electrostatic separation.

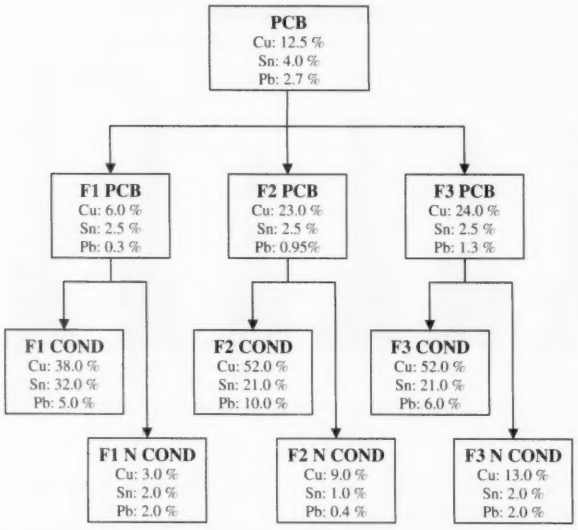

Fig. 6. Flowchart of enrichment/depletion of copper, tin and lead in the fractions as compared to the input.

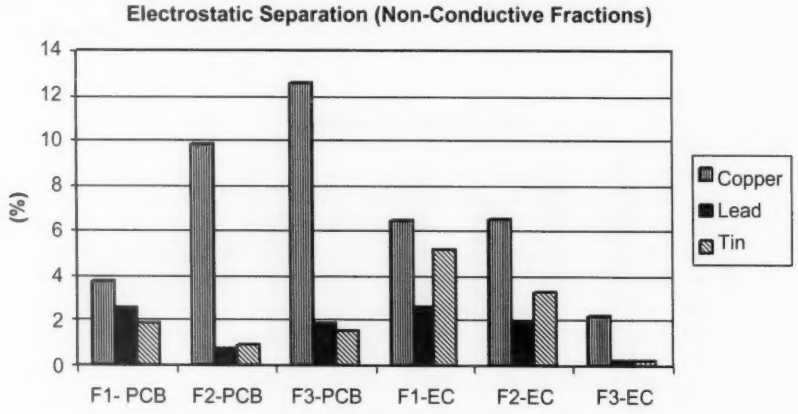

Fig. 7. Chemical analysis of the non-conductive fractions after electrostatic separation.

fraction (Fig. 4) and the contents of metals obtained after electrostatic separation (Figs. 5 and 6). The metals to be analyzed were chosen because they are good conductors and also because they are the metals with the highest content found in the printed circuit boards (compare Tables 1 and 2).

Fig. 5 presents the contents of Cu, Pb and Sn on the conductive fractions and Fig. 6 shows the chemical analysis of non-conductive fractions.

4. Discussion

It was expected that the fractions that were magnetically separated should have high concentrations of Fe, proving the efficiency of this type of separation.

Although the amount of magnetic material present in printed circuit boards is small, it is interesting to separate it previously, in order to obtain conductive fractions with higher copper content. The largest amount of magnetic material in whole printed circuit boards was obtained for the fraction F3: 54 g in 2 kg of samples. In electronic components 78 g in 2 kg was obtained for the fraction F3 (Fig. 2).

The concentration of iron in the magnetic fractions were, on average, 43% for whole printed circuit board and 46% for electronic components. The concentration of nickel was, on average, 15.2% for whole printed circuit board and 15.6% for electronic components (Fig. 3). Comparing these results with the ones shown in the Tables 1 and 2, it can be seen that there was a high concentration of iron and nickel after magnetic separation.

From the electrostatic separation it can be seen that the largest amount of conductive material was obtained for the fractions F3 of whole printed circuit board and F2 and F3 for electronic components (Fig. 2). The percentage of conducting material is quite significant, mainly in the fractions F2 and F3 of electronic components, with 25% on average of conducting material (Fig. 4).

It was expected to find the largest amount of magnetic and conductive materials in the fractions F3, because it is more difficult to mill metals than polymers and ceramics, therefore metals remain more concentrated in the largest fractions.

It can also be seen that the concentrations of copper reached more than 50% (in mass) in most of the conductive fractions and significant concentrations of Pb and Sn were present in all fractions (Fig. 5). Fig. 6 shows the enrichment/depletion of copper in the fractions as compared to the input. It can be seen that it was possible to concentrate copper, lead and tin after electrostatic separation when compared with initial percentages. The higher concentrations of these three elements after electrostatic separation was expected, because copper is a good conductor and it is present in large quantities in printed circuit boards. Pb and Sn are used in the welding of the electronic components to the substratum, in other words, they are also good conductors and are present in significant amounts. The same results can be observed in electronic components when Fig. 5, Tables 1 and 2 are compared.

The results shown in previous work (Veit et al., 2002b) about density separation of whole printed circuit boards and electronic components also show that copper, lead and tin are the main elements. The concentration of copper in the fractions after density separation (heavy fractions) was, on average, 55% for whole printed circuit board and 53% for electronic components. When we consider the lead content, there was 1.4% Pb in fractions after density separation for whole printed circuit board and 4% for electronic components. The same considerations done for tin showed an enrichment of 7% for whole printed circuit board and 11% for electronic components.

Comparing these results with the results shown in this paper of the fractions concentrated after magnetic and electrostatic separation, it can be seen that the concentrations of copper are similar, while lead and tin contents are higher at the conductive fraction (obtained by magnetic and electrostatic process) than at the dense fraction (obtained by density separation).

The conductive fractions, which are now concentrated in copper, will be treated by electrochemical techniques for the recovery of the copper by electrowinning.

The non-conductive fractions (polymers and ceramics) presented low contents of metals, thus proving the efficiency of the magnetic separation and the electrostatic separation. The exception is the contents of copper in F2 and F3 for whole printed circuit board (Fig. 7), which reached 9.8% and 12.5%, respectively. These contents probably would decrease if non-conductive material were treated again in the electrostatic separator. This will be tested in further works. The next steps will evaluate the energetic costs of the processes. In addition, the scale-up of the process will be studied in order to evaluate the possibilities of application on industrial scale.

5. Conclusions

The use of magnetic separation and electrostatic separation was efficient to obtain fractions with relatively high concentrations of metals starting from scrap of printed circuit boards. The fractions magnetically separated reached 43% of iron on average in whole printed circuit boards scrap (PCB) and 46% of iron in electronic components scrap (EC). The fractions electrostatically separated presented, for PCB, medium concentrations of 50% copper, 25% tin and 7% lead. For EC the concentrations were 46% copper, 23% tin and 8% lead.

Those fractions can now be used in secondary industries or applied as raw material in electrowinning techniques to separate most metals from each other.

The non-conductive fractions (that contain polymers and ceramics) need more studies to be suitably disposed of or to be sent to other recycling processes.

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Management of incinerator residues in Flanders (Belgium) and in neighbouring countries. A comparison

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Abstract

This paper covers the Flemish legislative tools concerning the management of bottom ash, fly ash and APC residue from municipal waste incinerators, with respect to their contamination with heavy metals. The situation in Flanders is compared to the one in the Walloon region, The Netherlands, Germany and France.

Waste management in the countries considered differs on the level of available management options, of leaching tests and of limit values. To make an indicative comparison of leaching tests and limit values in the different countries, leaching tests were carried out on bottom ash and fly ash, and the results are compared to the relevant limit values for recycling and landfilling of the different countries.

The comparison of legislations as well as the leaching results show that discrepancies in waste management between the different regions and countries exist. Recently, European limit values for landfilling became available. European legislation on recycling, however, has not been developed and urgently needs to be considered and drafted as the market for recycling can be expanding rapidly.
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1. Introduction

In a municipal solid waste incinerator (MSWI) different types of residues are produced (Fig. 1), the most important being bottom ash, which amounts to 20–30 wt% of the original municipal solid waste. Boiler ash is collected in the steam boiler. To clean the flue gases, usually reagents such as $NaHCO_3$ or $Ca(OH)_2$ are injected in a dry or semi-dry scrubber. Fly ash is collected separately by an electrostatic precipitator or along with the reaction products and excess reagent on a fabric filter. This may be followed by an additional wet flue gas cleaning. In this paper, fly ash is defined as solid residue originating from flue gas cleaning with no other products added; air pollution control (APC) residue is the flue gas cleaning residue together with reaction products and excess reagent. The ashes mentioned all contain heavy metals (IAWG, 1997), the most important being Cu, Pb and Zn (Table 1). Volatile metals such as Pb and Zn are usually found in higher concentrations in fly ash and APC residue than in bottom ash.

These residues are in practice recycled or landfilled. Advantages of recycling are that no landfill space is needed and that the materials may replace useful raw materials, which should be otherwise mined. Recycling may be stimulated by a landfill ban or by increasing landfill taxes. Fig. 2 illustrates the increase over the period 1989–2000 of the total cost of the landfilling of hazardous waste in Flanders (OVAM, 2001a). The total landfilling cost more than quadrupled, due to an in-

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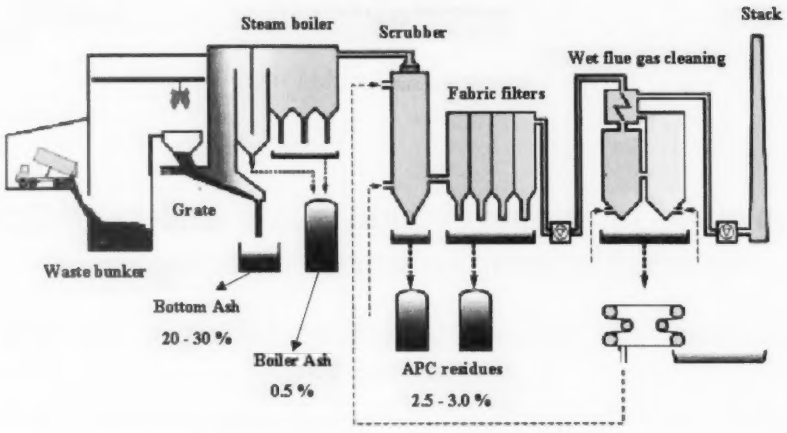

Fig. 1. Process scheme of a municipal solid waste incinerator.

crease of both the net price and the taxes. Recycling of incineration residues may be hampered due to considerations related to environmental safety and product quality.

In the first part of this paper, an overview is given of the legislation (both relative to recycling and landfilling) regarding incinerator residues in Flanders (Belgium) and in neighbouring regions and countries. Only inorganic compounds are included in the discussion. In the second part of this paper, the results of different leaching tests on bottom ash and fly ash are presented and compared to the relevant heavy metal leaching limit values of the different countries, in order to compare the different national legislations.

2. Overview of leaching tests

Leaching limit values and leaching results depend on the leaching test performed. The formally required leaching tests in the different countries are presented with focus on type, duration and *L/S* ratio. The type of leaching test refers to either column leaching with equilibrium not necessarily achieved or batch extraction test where equilibrium is assumed to have been reached.

Table 1
Heavy metal content of MSW residues (mg/kg dry matter) (IAWG, 1997)

	Bottom ash	Boiler ash	Fly ash	APC residue
As	1–200	20–60	40–300	20–500
Ba	400–2500	2000–3000	300–3000	50–14000
Cd	1–70	20–1300	50–450	140–1400
Co	20–50	20–200	10–90	1–300
Cr	20–3000	200–1000	140–1100	70–600
Cu	300–8000	500–1000	600–3200	10–2400
Mo	2–300	20–40	15–150	2–40
Ni	100–600	100–1500	60–260	20–700
Pb	100–14,000	1000–35,000	5300–26,000	2500–22,000
Sb	10–400	200–1000	260–1100	80–1100
Se	<10	10–100	1–30	1–30
Sn	2–400	200–700	550–2000	300–1400
Zn	1000–7000	5000–50,000	9000–70,000	7000–50,000

2.1. NEN7343 and prEN14405 column tests

For the NEN7343 test a column (20 cm length, 5 cm internal diameter) is filled with material, which was previously crushed to particle size <4 mm. The column is percolated with distilled water (acidified with concentrated HNO_3 down to pH 4) from bottom to top with a maximum water flow of 2 cm/h. The eluate is collected in several fractions depending on the total liquid to solid ratio (*L/S*) that needs to be reached. When the total *L/S* has to be 10, seven fractions are collected corresponding with *L/S* of 0.1, 0.1, 0.3, 0.5, 1, 3 and 5. The test takes about 3 weeks. When the total *L/S* has to be 1, only the first four fractions are collected. The leachate fractions are filtered over a 0.45 μm membrane and conserved for analysis. The metal concentrations in the consecutive fractions are summed to obtain a cumulative leached concentration.

The draft prEN14405 procedure is similar to the NEN7343 column test but the total *L/S* is 0.1. The single leachate fraction of the EN test equals in fact the first fraction of the NEN test.

2.2. DIN38414-S4, EN12457/4 and NFX31-210 extraction tests

In the DIN test, 20 g of material crushed to particles <4 mm, is leached in 200 ml of distilled water (*L/S* = 10) for 24 h while agitating. The slurry is then filtered over a 0.45 μm membrane to give a leachate, which can be analysed.

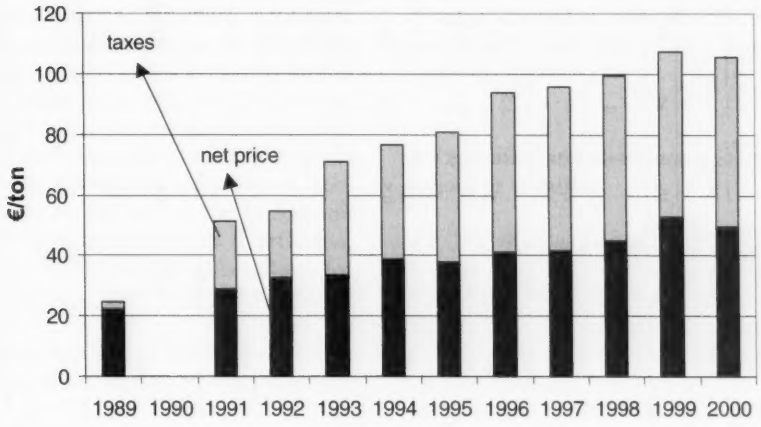

Fig. 2. Evolution of landfill cost for hazardous waste in Flanders.

The EN procedure is similar to the DIN test.

In the NFX test, the material is leached for only 16 h, but the extraction is repeated two times reusing the residue on the 0.45 μm filter membrane ($L/S = 30$). The metal concentrations in the three leachates are summed.

3. Legislation and waste management in Flanders

In Flanders (population = 5.95 million), 67% of MSW was collected separately in 2001 (VMM, 2002). Eleven MSW-incinerators incinerated in 1999 ca. 900,000 tons of solid waste. This led to 200,000 tons of bottom ash and to 35,000 tons of APC residue and boiler ash (OVAM, 2001b). In 2001, 258,000 tons of bottom ash were produced (OVAM, 2003).

3.1. Recycling

The Flemish government has drafted a list of hazardous wastes according to European legislation. Bottom ash is not hazardous and can, from the legal point of view, be considered for recycling in construction applications on condition that the application complies with certain requirements. For inorganic compounds (e.g., heavy metals), limit values were defined in Flanders for the leaching concentration of granular material (Table 2), using the NEN7343 column test. These limit values are valid for granular materials with a density of 1550 kg/m^3 and which are applied in a volume with a height of 0.7 m. In other conditions, laboratory leaching results should be recalculated to immission values and compared to immission limit values for soil. When granular materials comply with the limit values, they can be used as such or as an aggregate in monolithic applications. Granular materials that do not comply with the limit leaching values for granular applications can be recycled in monolithic applications, provided that the immission concentrations, calculated from the laboratory results using the NEN7345 diffusion test, comply with the immission limit values for soil. Besides the environmental limit values, civil-technical requirements (grain size distribution, strength, etc.) have to be met as well. When both leaching and civil-technical limit values are complied with, utilisation is unrestricted with respect to isolation measures.

In practice, immediate recycling of MSWI-bottom ashes is not possible in Flanders, because the leaching limit values are exceeded for copper and occasionally for lead and zinc (OVAM, 2001b; Vandecasteele et al., 2002). Also the particle size distribution of bottom ashes does not comply with that specified in the Belgian technical description of building materials, which applies to all construction activities for the government. So bottom ashes either have to be treated before recycling or have to be landfilled.

Unlike the situation in The Netherlands and Germany, treatment of bottom ashes, such as magnetic separation of iron, non-ferro separation, sieving and crushing of particles >40 mm, is not standard procedure. In Flanders two main bottom ash treatment plants are in operation (OVAM, 2001b). One is a wet sieving system with a capacity of 165,000 tons/year and the other a dry sieving system with a capacity of 300,000 tons/year. The wet processing of bottom ashes has several clear advantages over the dry system in view of quality-assurance: the light organic fraction floats in the washing barrel and can easily be removed, soluble salts are readily washed out, the rinsing effect produces visual clean end-products, and a high sieving efficiency is obtained producing high quality granulates. Bottom ashes processed by the wet sieving system are separated into ferrous and non-ferrous fractions which can be recycled, a 0–0.1 mm sludge fraction which has to be landfilled, a 0.1–2 mm sand fraction which can be used as construction material (e.g., on landfills), and 2–6

Table 2
Limit values for recycling of waste as construction material in selected countries

Category	Flanders		Wallonia		The Netherlands			Germany									France	
	All	Granular	BA		Cat. 1	Cat. 2	BA	Z_0		$Z_{1.1}$		$Z_{1.2}$		Z_2		BA	V	M
Criterion	Content[a]	Leaching	Leaching		Leaching[c]			Content	Leaching	Content	Leaching	Content	Leaching	Content	Leaching	Leaching	Leaching	
Test	Destr.[b]	NEN column	DIN[d] extr.	NEN column	NEN column			Destr.[b]	DIN[d] extr.	Destr.[b]	[d] extr.	Destr.[b]	[d] extr.	Destr.[b]	DIN[d] extr.	DIN[d] extr.	NFX extr.	
L/S	–	10	10	10	10	10	10	–	10	–	10	–	10	–	10	10	30	30
Al	–	–	2000	2000	–	–	–	–	–	–	–	–	–	–	–	–	–	–
As	250	0.8	1.0	0.8	0.88	7.0	7.0	20	0.1	30	0.1	50	0.4	150	0.6	–	2	4
Ba	–	–	–	–	5.5	58	58	–	–	–	–	–	–	–	–	–	–	–
Cd	10	0.03	1.0[e]	0.03	0.032	0.066	0.066	0.6	0.02	1	0.02	3	0.05	10	0.1	0.05	1	2
Co	–	–	1.0	0.25	0.42	2.5	2.5	–	–	–	–	–	–	–	–	–	–	–
Cr	1250	0.5	–	0.5	1.3	12	12	50	0.15	100	0.3	200	0.75	600	1.5	2	–	–
Cr^{VI}	–	–	1.0[e]	0.05	–	–	–	–	–	–	–	–	–	–	–	–	1.5	3
Cu	375	0.5	20[e]	5.0	0.72	3.5	24	40	0.5	100	0.5	200	1.5	600	3	3	–	–
Hg	5	0.02	0.2[e]	0.02	0.018	0.076	0.076	0.3	0.002	1	0.002	3	0.01	10	0.02	0.01	0.2	0.4
K	–	–	–	1700	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Mo	–	–	1.5	1.8	0.28	0.91	26	–	–	–	–	–	–	–	–	–	–	–
Ni	250	0.75	2.0[e]	1.8	1.1	3.7	3.7	40	0.4	100	0.5	200	1.5	600	2	0.4	–	–
Pb	1250	1.3	2.0[e]	2.2	1.9	8.7	8.7	100	0.2	200	0.4	300	1	1000	2	0.5	10	50
Sb	–	–	2.0	0.3	0.045	0.43	2.1	–	–	–	–	–	–	–	–	–	–	–
Se	–	–	–	–	0.044	0.1	0.1	–	–	–	–	–	–	–	–	–	–	–
Sn	–	–	–	–	0.27	2.4	2.4	–	–	–	–	–	–	–	–	–	–	–
V	–	–	–	–	1.6	32	32	–	–	–	–	–	–	–	–	–	–	–
Th	–	–	–	–	–	–	–	0.5	<0.01	1	0.01	2	0.03	10	0.05	–	–	–
Ti	–	–	20	2.4	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Zn	1250	2.8	9.0[e]	4.0	3.8	15	15	120	1	300	1	500	3	1500	6	3	–	–
Br^-	–	–	–	–	2.9[f]	44[f]	44[f]	–	–	–	–	–	–	–	–	–	–	–
Cl^-	–	–	5000	6000	599[f]	8807[f]	8807[f]	–	100	–	100	–	200	–	300	2500	–	–
F^-	–	–	50	20	13[f]	102[f]	102[f]	–	–	–	–	–	–	–	–	–	–	–
SO_4^{2-}	–	–	10,000	4000	1136[f]	22,027[f]	22,027[f]	–	500	–	500	–	1000	–	1500	6000	10,000	15,000
Total CN^-	–	–	0.46	0.2	–	–	–	1	<0.1	10	0.1	30	0.5	100	1	–	–	–
Free CN^-	–	–	–	–	–	–	–	1	–	10	–	20	–	100	–	0.2	–	–
pH	–	–	7–12	–	–	–	–	5.5–8[a]	5.5–10[a]	5.5–8[a]	5.5–12[a]	5–9[a]	5.5–12[a]	–	–	7–13	–	–

All values are expressed in mg/kg dry matter for the sake of comparison, although formal values are sometimes in mg/l.

[a] Indicative values.

[b] Destruction in Flanders has to be performed with HCl, HNO_3 and HF; destruction in Germany is performed with Aqua Regia.

[c] The leaching values for granular material in The Netherlands are calculated from the immission limit values for judgment purposes, using default values in the conversion formula as proposed by KIWA (1999) and using the same parameters as in Flanders (material density = 1550 kg/m^3, height of used volume = 0.7 m).

[d] Formal limit values are expressed in mg/l but are recalculated to mg/kg by multiplication with the applied *L/S* ratio.

[e] Sum of Cd, Cr^{VI}, Cu, Hg, Ni, Pb and Zn has to be lower than 50 mg/kg dry matter.

[f] Other limit values apply for specific conditions.

and 6–50 mm granulate fractions for which certificates were obtained for use as granular material in foundations. The granulates have to be stored for ageing during three months prior to application. For the 2–6 mm fraction, a certificate was obtained for use as replacement of raw materials such as sand and gravel in concrete bricks (Jaspers, 2002). The dry sieving system produces fractions of 0–10, 10–22, 22–40 and 40–80 mm in addition to the ferrous and non-ferrous fractions (Cortvriend, 2001). The fractions of 10–40 mm have a certificate for use in underfoundation, but only for specific applications.

To date, a significant fraction of the treated bottom ashes in Flanders is recycled, mostly in Wallonia. The market in Flanders itself is still immature. This can partly be explained by the very low limit value for Cu in comparison with Wallonia and neighbouring countries and by the only recent emergence of the wet sieving technology. In time this market is expected to grow substantially (Vrancken et al., 2000). In 2001, 54,000 tons of bottom ashes were exported to The Netherlands and 17,000 tons to Germany (OVAM, 2003).

3.2. Landfilling

When incinerator waste cannot be recycled, landfilling is the only option. The criteria for disposal of wastes are specified in the legislation. Fly ash and APC residue from waste incineration are hazardous wastes and have to be disposed of in category-1 landfills, which are landfills for the disposal of non-reactive hazardous wastes and for non-hazardous wastes of mainly inorganic composition. The leachability of heavy metals, following the DIN38414-S4 leaching test procedure, has to comply with the limit values given in Table 3. If the waste does not comply with these leaching limit values, immobilisation of the waste with additives such as cement has to be applied. If the best available technology for immobilisation does not produce a result that complies with all requirements, so-called "salt cell" conditions should be used in which a physical barrier prevents contact of the waste with percolating water. Unlike fly ash and APC residue, bottom ash from municipal waste incineration is also accepted in category-2 landfills, which are exclusively for non-hazardous wastes. The leaching criteria are similar to those for category-1 landfills.

No fly ash or APC residue from waste incineration is recycled in Flanders. These residues are landfilled in category-1 landfills. The soluble salt content and the lead leachability exceed the limit values for disposal in category-1 landfills; therefore, fly ash and APC residue have to be treated before disposal. Adding cement is a common technique but cannot always reduce Pb leaching to below the limit value. Other immobilisation recipes then have to be applied. These additives, however, do not reduce the soluble salt content to below the 10% limit value. So-called salt cell conditions are therefore needed to prevent contact with percolating water (Geysen et al., 2004a,b).

4. Legislation and waste management in neighbouring regions or countries

4.1. The Walloon region

In 2001 Wallonia (population = 3.35 million) collected 53% of its MSW selectively, which leaves 650,000 tons of solid waste to be terminally removed. Of this amount, 28% was incinerated in 4 incinerator installations, resulting in about 37,000 tons of bottom ash (CEE, 2003).

4.1.1. Recycling

In Wallonia bottom ash, fly ash and APC residue can be labeled as either hazardous or non-hazardous, depending on the total content of, among others, heavy metals. In practice, bottom ash is assumed to be non-hazardous, while fly ash and APC residue are considered possibly hazardous. The latter are therefore not included in the list of wastes that are eligible for recycling. Bottom ash does appear on this list and can be recycled as such in foundation constructions or as an aggregate in concrete. However, of all materials on the list, bottom ash is the most rigorously tested. Leaching of either application (as such or as aggregate in a monolith) has to comply with a set of limit values using the NEN7343 column test and with a set of limit values using the DIN38414-S4 extraction test (Table 2). These limit values are specifically to be used in the case of bottom ash.

4.1.2. Landfilling

The Walloon region has established three main classes of landfills: class-1 landfill accepts hazardous wastes, class-2 is for the disposal of non-hazardous industrial and municipal waste, and class-3 for inert waste. Besides these categories, landfills exist for dredging sludge (class-4) and for exclusive use by one waste producer (class-5). To date no limit values exist as acceptance criteria for waste. Acceptance is based on the broad definitions of hazardous, non-hazardous and inert waste. Fly ash and APC residue should be landfilled in a class-1 landfill. Wallonia, however, does not have such a landfill (CEE, 2003). These residues are then either transported abroad or treated in order to landfill them in class-2 landfills (MRW, 1998).

4.2. The Netherlands

In the Netherlands (population = 16 million), 11 large MSW-incinerators incinerate about 5 million tons

Table 3
Limit values for disposal of hazardous (H) and non-hazardous (NH) waste in selected countries

Category	Flanders		The Netherlands	Germany			France			European Union[a]					
	II	I	C_3	Z_3	Z_4	Z_5	I	IA	IB	Non-hazardous			Hazardous		
Waste	NH	NH/H	H	NH	NH	H	H	H	H	NH/H			H		
Test	DIN[b] extr.	DIN[b] extr.	NEN column	DIN[b] extr.	DIN[b] extr.	DIN[b] extr.	NFX extr.	NFX extr.	NFX extr.	EN extr.	EN extr.	EN[b] column	EN extr.	EN extr.	EN[b] column
L/S	10	10	1	10	10	10	30	30	30	2	10	0.1	2	10	0.1
As	10	10	9	2	5	10	10	30	30	0.4	2	3	6	25	30
Ba	–	–	60	–	–	–	–	–	–	30	100	200	100	300	600
Cd	5	5	0.2	0.5	1	5	25	100	50	0.6	1	3	3	5	17
Co	–	–	6	–	–	–	–	–	–	–	–	–	–	–	–
Cr	–	–	30	–	–	–	50	100	100	4	10	25	25	70	150
Cr^{VI}	5	5	–	0.5	1	5	5	30	20	–	–	–	–	–	–
Cu	100	100	10	10	50	100	–	–	–	25	50	300	50	100	600
Hg	1	1	0.1	0.05	0.2	1	5	10	10	0.05	0.2	0.3	0.5	2	3
Mo	–	–	3	–	–	–	–	–	–	5	10	35	20	30	100
Ni	20	20	10	2	10	20	50	100	100	5	10	30	20	40	120
Pb	20	20	25	2	10	20	50	2000	100	5	10	30	25	50	150
Sb	–	–	0.8	–	–	–	–	–	–	0.2	0.7	1.5	2	5	10
Se	–	–	0.3	–	–	–	–	–	–	0.3	0.5	2	4	7	30
Sn	–	–	6	–	–	–	–	–	–	–	–	–	–	–	–
V	–	–	20	–	–	–	–	–	–	–	–	–	–	–	–
W	–	–	0.5	–	–	–	–	–	–	–	–	–	–	–	–
Zn	100	100	40	20	50	100	250	500	500	25	50	150	90	200	600
Br^-	–	–	160	–	–	–	–	–	–	–	–	–	–	–	–
Cl^-	10,000	–	50,000	–	–	100,000	–	–	–	10,000	15,000	85,000	17,000	25,000	150,000
F^-	500	500[c]	280	50	250	500	–	–	–	60	150	400	200	500	1200
SO_4^{2-}	10,000	–	80,000	–	–	50,000	–	–	–	10,000	20,000	70,000	25,000	50,000	170,000
CN^-	10	10	5	–	–	–	5	10	10	–	–	–	–	–	–
Free CN^-	–	–	3	1	5	10	–	–	–	–	–	–	–	–	–
NO_2^-	300	300		–	–	300	–	–	–	–	–	–	–	–	–
NH_4^+	–	10,000[c]		40	250	10,000	–	–	–	–	–	–	–	–	–
pH	–	4–13	3–13	5.5–13	5.5–13	4–13	4–13	4–13	4–13	>6	>6	>6	–	–	–

All values are expressed in mg/kg dry matter for the sake of comparison, although formal values are sometimes in mg/l.

[a] The limit values given in European legislation should be consulted together with the many exceptions that are made possible in the legislative articles.

[b] Formal limit values are expressed in mg/l but are recalculated to mg/kg by multiplication with the applied *L/S* ratio.

[c] Indicative value.

of solid waste per year. This leads to more than 1 million tons of bottom ash and 150,000 tons of APC residue and boiler ash (Lamers and van Aalten, 2001).

4.2.1. Recycling

It is the Dutch aim to responsibly maximize recycling of MSWI residues and avoid landfilling as much as possible. The Netherlands are pioneers in recycling waste for construction applications. Waste can be recycled in granular or in monolithic applications. Waste can be applied as construction material when immission into the soil of selected compounds is below limit values (Table 2). The immission values have to be calculated from test results obtained with the NEN7345 diffusion test for monolithic applications and with the NEN7343 column test for granular applications. The result of this calculation depends on where and how much of the material is applied. When the material complies with the immission limit values for category-1 construction material, it is classified as such and no isolation precautions are needed to prevent contact with water. When the immission limit values for category-1 are exceeded, but category-2 limit values are complied with, the waste can be recycled as category-2 construction material provided all necessary isolation measures are taken.

Bottom ash from waste incineration does not, even after isolation measures, comply with the immission limit values for copper, molybdenum and antimony (KIWA, 1999) and thus does not comply with the demands for category-2 construction material. Since large-scale landfilling of bottom ashes is an undesirable option in Dutch policy, the MSWI sector has been summoned to improve the quality of the residues through upgrading. As development and implementation of upgrading techniques will take time, a special category with adjusted, less stringent limit values is temporarily created so that these bottom ashes can in the intermediate period be recycled (BA in Table 2). Extra isolation measures have to be taken in comparison with category-2 construction material. This special category for bottom ashes will cease to exist on 31 December 2005. When the bottom ash complies with the requirements of the special category, a recycling certificate is delivered. This implies that a guideline for recycling of bottom ashes with recycling applications and control protocols has to be followed.

Until 2002 all bottom ashes from waste incineration have been recycled as foundations or as embankment for road or railway construction (Lamers and van Aalten, 2001). Bottom ashes are sieved in fractions <10–12, <40 and >40 mm. The fraction >40 mm is crushed and sieved again. Iron scrap and non-ferrous materials are removed. The different fractions have to age for at least six weeks before use. In 2000, 280,000 tons of treated bottom ashes complied with the requirements for category-2 construction material, and 630,000 tons were considered as special category MSWI-bottom ashes. Due to the number of large-scale constructions in which bottom ash can be utilized, the demand in the years 2000–2001 has been such that even bottom ash from Belgium was accepted for treatment and used in The Netherlands for construction applications.

4.2.2. Landfilling

Recycling of fly ash and APC residue is not possible because limit values are exceeded even with isolation measures; these incinerator wastes have to be landfilled. Hazardous wastes have to be landfilled on a C_2 or C_3 landfill. The selection of the landfill category depends on the leachability of several compounds following the short NEN7343 column leaching protocol with $L/S = 1$. Limit values are given in Table 3. If concentrations in the leachate are below the limit values, the waste is categorized as C_3. If one or more of the leaching values are above or equal to the corresponding limit values, the waste has to be categorized as C_2. C_1 waste is a collection of very specific hazardous wastes, e.g. Hg-containing waste, untreated As_2S_3 sludge, etc., for which The Netherlands does not have landfilling capacity. In The Netherlands there is one landfill specifically for C_2 wastes, which is a concrete bunker with a capacity of 230,000 tons hazardous waste, and several C_3 landfills.

Since 1999 landfilling of fly ash and APC residue without prior treatment has been prohibited. These wastes can be stored in C_3 landfills in compartments that are made suitable for accepting C_2 wastes or in compartments that are made suitable for immobilized wastes. In addition, one-fifth of the fly ash from waste incineration is recycled as asphalt filler (containing 25% of fly ash). Besides landfilling and recycling as filler in asphalt, fly ash is also exported from The Netherlands to Germany and dumped there as stabilization material for preventing the collapse of salt mines and for filling up old coalmines.

4.3. Germany

4.3.1. Recycling

In Germany, wastes are divided in 7 categories depending on the composition and the leachability according to the DIN38414-S4 leaching protocol. The categories are Z_0, $Z_{1.1}$, $Z_{1.2}$, Z_2, Z_3, Z_4 and Z_5. The limit values are given in Tables 2 and 3. Z_0 wastes can be recycled as construction material without any limitation. $Z_{1.1}$, $Z_{1.2}$ and Z_2 waste have more limited use: $Z_{1.1}$ can be used in hydrologically unfavourable conditions, $Z_{1.2}$ in hydrologically favourable conditions and for Z_2 wastes, technical protection measures have to be taken.

Bottom ashes can be recycled for road construction. Besides the above-mentioned categories, a special category for bottom ashes has been created (BA in Table 2), since leaching of salts from MSWI ashes cannot

comply with Z_2 limit values. When recycling is not possible, the bottom ashes should be landfilled as non-hazardous waste.

4.3.2. Landfilling

Z_3 wastes must be landfilled in category-1 landfills, Z_4 waste in category-2 landfills and Z_5 wastes in landfills for hazardous materials. Both category-1 and -2 landfills are for non-hazardous waste, but category 2 is for wastes that have a higher leachability than wastes in category 1. Landfills for hazardous waste can be above ground or underground in salt mines. When hazardous wastes do not comply with the Z_5 limit values, they have to be treated or stored in underground salt mines in big bags.

The German legislation has identified refilling old mines as a useful application for fly ashes and APC residues. This approach has been criticized, because the long-term consequences are less clear (Gebers and Küppers, 1998; Schmitz and Wermuth, 2001).

4.4. France

4.4.1. Recycling

Three types of recycling are distinguished, based on the NFX31-210 extraction test (Table 2). Bottom ash with a low leachability is of the V-type (valorization) and can be recycled immediately in road and other constructions without taking isolation measures. M-type bottom ash (maturation) has a medium leachability and needs treatment before recycling. This treatment can be solidification with a hydraulic binder such as cement, or ageing. Without treatment this waste has to be landfilled in a category-II landfill. Bottom ash with a too high level of leachability belongs to the S-type (storage) and has to be landfilled in a category-II landfill with prior stabilization if needed. Limit values are given for only six elements.

4.4.2. Landfilling

Incinerator residues that are not recycled, are to be landfilled in a category-I or -II landfill. Category-I landfills are for hazardous inorganic industrial waste, category-II landfills for non-hazardous organic waste. Industrial waste that has to be stabilized in order to comply with the leaching limit values for a category-I landfill (Table 3) can be temporarily stored before treatment. Depending on their leachability, which is to be tested with the NFX31-210 extraction test, these stored wastes have to be treated within two (category IA) or five (category IB) years. The limit values for identifying waste as IA or IB depend on the type of waste and are given for incinerator residues in Table 3. Fly ash and APC residue have to be landfilled in a category-I landfill. Non-recyclable bottom ash has to be landfilled in a category-II landfill, more specifically in a category-IIE2. Other types of category-II landfills are for other wastes like plastic, metal and glass (IIE1), other mineral wastes (IIE3), etc.

5. European legislation

The recycling of incinerator residues is not yet regulated at the European level.

The European Union however has recently developed and implemented a policy on landfilling of wastes. Two landfill types are applicable to incinerator residues. On the one hand, there are landfills for non-hazardous wastes including municipal waste and other non-hazardous waste. Also stable non-reactive hazardous waste (e.g., solidified waste) that complies with the acceptance criteria of the landfill can be disposed of in this landfill. On the other hand there are landfills or underground storage facilities for hazardous wastes. The European Directive, which came into effect in July 1999, establishes licensing criteria of landfills and acceptance procedures for accepting waste at these landfills. Leaching criteria were published in a Council Decision of January 2003, taking effect on July 16, 2004. The leaching criteria however will only take effect a year later on July 16, 2005. Table 3 shows the limit values for the two types of landfill. These values are for granular waste. The member states should establish criteria for monolithic waste to provide the same level of environmental protection given by the limit values for granular waste. The European Union also leaves it up to the member states to decide which of the three permitted leaching tests are used: an extraction test with $L/S = 2$ (EN12457-1/3), an extraction test with $L/S = 10$ (EN12457-2/4) or a short column test with $L/S = 0.1$ (prEN14405). Instead of the prEN14405 draft standard procedure, member states can also use national procedures approved by the European authorities.

6. Practical evaluation of a bottom ash and fly ash in view of recycling and disposal

6.1. Materials and methods

A treated bottom ash fraction from a municipal solid waste incinerator in Flanders was chosen for its high leachability of several heavy metals and was tested for its recycling possibilities as a granular material. A fly ash from an electrostatic precipitator in an industrial waste incinerator in Flanders was tested for its compliance with the landfill limit values of the different national legislations. The fly ash was immobilized with up to 40% of cement (CEM I 42.5 LA) and leached after 7 days of hardening.

Table 4
Results of column tests (mg/kg dry matter, average of 2 tests) of a bottom ash fraction compared to limit values in Flanders, Wallonia and The Netherlands (total $L/S = 10$)

	Experiment	Flanders granular	Wallonia BA	The Netherlands		
				Cat. 1	Cat. 2	BA
As	0.20	0.8	0.8	0.88	7.0	7.0
Ba	n.a.	–	–	5.5	58	58
Cd	0.01	0.03	0.03	0.032	0.066	0.066
Co	n.a.	–	0.25	0.42	2.5	2.5
Cr	0.08	0.5	0.5	1.3	12	12
Cu	**0.63**	**0.5**	5.0	0.72	3.5	24
Hg	n.a.	0.02	0.02	0.018	0.076	0.076
Mo	**0.51**	–	1.8	**0.28**	0.91	26
Ni	0.01	0.75	1.8	1.1	3.7	3.7
Pb	0.13	1.3	2.2	1.9	8.7	8.7
Sb	**0.79**	–	**0.3**	**0.045**	**0.43**	2.1
Se	n.a.	–	–	0.044	0.1	0.1
Sn	n.a.	–	–	0.27	2.4	2.4
V	n.a.	–	–	1.6	32	32
Zn	1.31	2.8	4.0	3.8	15	15
pH	8.2–9.6	–	–	–	–	–

n.a. = not analyzed.

Leaching was performed as described in an earlier paragraph. All leachates were acidified with concentrated HNO_3 and subsequently analyzed for metal concentrations using a VG PlasmaQuad PQ-2 Plus ICP-mass spectrometer.

6.2. Results and discussion

6.2.1. Leaching experiments for recycling of a bottom ash fraction

Recycling of the bottom ash fraction was tested with the NEN7343 column test (with a total $L/S = 10$) for comparison with limit values in Flanders, Wallonia and The Netherlands and with the DIN and NFX extraction tests for comparison with limit values in Wallonia, Germany and France.

Table 6
Results of NFX extraction tests (mg/kg dry matter, average of 2 tests) of a bottom ash fraction compared to limit values in France

	Experiment	V	M
As	n.a.	2.0	4.0
Cd	0.3	1.0	2.0
Cr^{VI}	0.1[a]	1.5	3.0
Hg	n.a.	0.2	0.4
Pb	0.2	10	50
pH	10.0–10.3	–	–

n.a. = not analyzed.
[a] This is total Cr.

The results are summarized in Tables 4–6. Leaching exceeded the Flemish limit value for Cu (Table 4). Ageing of the bottom ash fraction for three months decreases Cu leaching to below the limit value (Jaspers, 2002; Vandecasteele et al., 2002). In The Netherlands, this bottom ash fraction would exceed the leaching limit values for Category I in the case of Mo and Sb and for Category II in the case of Sb. There would be no problem for recycling in the special category for bottom ashes. In Wallonia, there is only a problem with Sb in the column test. In the extraction test there is no exceedance (Table 5). However, the results for both the column test and the extraction test need to comply with the limit values. The bottom ash fraction is not suitable for recycling in Germany: the limit values are exceeded for Z_0 with unrestricted reuse (As, Cd, Cr and Pb), $Z_{1.1}$, which allows reuse in hydrologically unfavourable conditions (As, Cd and Cr), $Z_{1.2}$ allowing reutilization in hydrologically favourable conditions (As and Cd), Z_2 where isolation measures need to be taken (As and Cd) and even for the special category for bottom ashes (Cd). In France immediate recycling is possible (Table 6).

The results of the column test and the two extraction tests differ substantially. This is mainly due to the difference in pH and whether or not agitation is applied.

Table 5
Results of DIN extraction tests (mg/kg dry matter, average of 2 tests) of a bottom ash fraction compared to limit values in Wallonia and Germany

	Experiment	Wallonia BA	Germany				
			Z_0	$Z_{1.1}$	$Z_{1.2}$	Z_2	BA
As	**0.8**	1.0	**0.1**	**0.1**	**0.4**	**0.6**	–
Cd	**0.3**	1.0	**0.02**	**0.02**	**0.05**	**0.1**	**0.05**
Cr	**0.7**	1.0[a]	**0.15**	**0.3**	0.75	1.5	2
Cu	0.5	20	0.5	0.5	1.5	3	3
Hg	n.a.	0.2	0.002	0.002	0.01	0.02	0.01
Ni	<0.1	2.0	0.4	0.5	1.5	2	0.4
Pb	**0.3**	2.0	**0.2**	0.4	1	2	0.5
Th	n.a.	–	<0.01	0.01	0.03	0.05	–
Zn	0.1	9.0	1	1	3	6	3
pH	10.2	7–12	5.5–10[b]	5.5–12[b]	5.5–12[b]	–	7–13

n.a. = not analyzed.
[a] This is Cr^{VI}.
[b] Indicative values.

Table 7
Results of DIN extraction tests (mg/kg dry matter) of a fly ash compared to limit values in Flanders, Germany and the EU

	Experiment					Flanders	Germany	EU
	No cement[a]	+10% cement	+20% cement	+30% cement	+40% cement	Cat. I landfill	Z_5	Hazardous waste
As	4	2	03	0.4	0.2	10	10	25
Cd	**163**	**49**	0.1	0.1	<0.1	**5**	**5**	**5**
Cr	1	0.4	5	2	2	5[b]	5[b]	70
Cu	56	36	23	23	5	100	100	100
Hg	n.a.	n.a.	n.a.	n.a.	n.a.	1	1	2
Ni	14	2	<0.1	<0.1	<0.1	20	20	40
Pb	**32**	16	1	0.3	0.1	**20**	**20**	**50**
Zn	**22,000**	**420**	64	63	26	**100**	**100**	**200**
pH	6.8	8.1	11.6	12.1	12.4	4–13	4–13	–

n.a. = not analyzed.
[a] Average of 2 tests.
[b] This is Cr^{VI}.

6.2.2. Leaching experiments for landfilling of a fly ash

To evaluate whether the fly ash can be landfilled immediately or has to be stabilized, the DIN extraction test was carried out for comparison with limit values for hazardous waste in Flanders, Germany and the EU, the NEN7343 column test was used for comparison with the Dutch (total $L/S = 1$) and European (total $L/S = 0.1$) limit values and the NFX31-210 extraction test was used for comparison with French limit values.

The results are shown in Tables 7–9. The Flemish limit values for landfilling in a category-2 landfill are the same as the German Z_5 limit values for landfilling of hazardous waste. Without stabilization the fly ash cannot be landfilled as the limit values are exceeded for Cd, Pb and Zn (Table 7). In Germany this fly ash could be disposed of in salt mines stored in big bags. Addition of 20% cement is needed for compliance with the above-mentioned limit values. In The Netherlands the fly ash does not comply with the requirements of a C_3 landfill (the limit values for Cd, Co, Cu and Zn are exceeded) and has to be landfilled in a C_2 landfill or in a special compartment of a C_3 landfill (Table 8). In France Cd and Zn leaching exceeds the limit value for landfilling on a category-I landfill (Table 9). Due to the high leaching of Cd, Pb and Zn, no temporary storage of the waste before treatment can be permitted. Addition of cement as a hydraulic binder does not decrease leaching of Zn below the limit value of a category-I landfill. Other additives or techniques have to be investigated. When comparing results with the European limit values, leaching exceeds the limit values for Cd and Zn both in the extraction test (Table 7) and the column test (Table 8). Using the extraction test, 20% of cement has to be added to the waste in order to make it acceptable for disposal in a landfill for hazardous waste. This corresponds with the required treatment in Flanders and Germany.

Table 8
Results of column tests of a fly ash (mg/kg dry matter, average of 2 results) compared to the Dutch and the European limit values

	The Netherlands ($L/S = 1$)		European Union ($L/S = 0.1$)	
	Experiment	C_3-limit value	Experiment	Hazardous
As	1.3	9	23	30
Ba	n.a.	60	n.a.	600
Cd	**138**	**0.2**	**2140**	**17**
Co	**6.5**	**6**	69	–
Cr	0.8	30	16	150
Cu	**12.4**	**10**	140	600
Hg	n.a.	0.1	n.a.	3
Mo	0.6	3	8	100
Ni	7.5	10	80	120
Pb	2.6	25	42	150
Sb	0.2	0.8	3	10
Se	n.a.	0.3	n.a.	30
Sn	n.a.	6	n.a.	–
V	n.a.	20	n.a.	–
W	n.a.	0.5	n.a.	–
Zn	**9466**	**40**	**116,300**	**600**
pH	5.6–6.1	3–13	5.7	–

n.a. = not analyzed.

7. Comparison of waste management under the different legislations

Waste management in the countries considered differs at three levels. At the level of *management options*, all countries distinguish between landfilling and recycling of waste.

In Wallonia, The Netherlands, Germany and France recycling of bottom ash has been specifically regulated. Limit vales of some components are less stringent compared to the limit values required for recycling of other waste types (e.g., Cu, Mo and Sb in The Netherlands; As, Cr, Th, Cl^-, SO_4^{2-}, total CN^- in Germany), while limit values of other components have been tightened (e.g., Cd, Hg, Ni, Pb, Zn, free CN^- in Germany) in order to facilitate reuse of bottom ash. Additional precautionary measures also have to be taken. In Flanders no

Table 9
Results of NFX extraction tests (mg/kg dry matter) of a fly ash compared to French limit values

	Experiment					Landfill	Storage	
	No cement[a]	+10% cement	+20% cement	+30% cement	+40% cement	Cat. I	Cat. IA	Cat. IB
As	4.4	5.4	2.3	5.1	1.9	10	30	30
Cd	**186**	**71**	0.1	1.3	1.2	**25**	*100*	*50*
Cr	0.6	7.6	8.2	13.3	13.6	50	100	100
Cr^{VI}	n.a.	n.a.	n.a.	n.a.	n.a.	5	30	20
Hg	n.a.	n.a.	n.a.	n.a.	n.a.	5	10	10
Ni	16.1	5.6	2.5	0.4	0.2	50	100	100
Pb	**58**	31	10.9	50	**76**	**50**	2000	100
Zn	**24,771**	**5376**	**848**	**432**	**460**	**250**	*500*	*500*
pH	6.7–8.5	7.2–11.1	11.1–11.5	11.8–11.9	11.9–12.2	4–13	4–13	4–13

n.a. = not analyzed.
[a] Average of 2 tests.

specific limit values exist for bottom ash. With the very low limit value for Cu leaching, the Flemish market for bottom ash recycling is severely hampered. All countries are less inclined to recycle fly ash and APC residue than bottom ash, but The Netherlands and Germany recycle a fraction of these wastes: as filler in asphalt (The Netherlands) or as stabilization material for salt mines (Germany).

Within the landfill option, more similarities exist. All countries consider bottom ash as non-hazardous waste whereas fly ash and APC residue are considered hazardous, so that these must be disposed of in different landfills.

With regard to *leaching tests*, column tests and extraction tests are applied. While The Netherlands use a column test to test landfilling and recycling, Germany and France use an extraction test for landfilling and recycling. Flanders makes use of a column test for recycling and an extraction test for landfilling. Wallonia requires testing by means of both the column test and the extraction test for recycling options; no leaching tests are required for landfilling. The European legislation permits extraction tests as well as a short column test for landfilling as a compromise.

As to the *limit values*, limit values are available for more metals in The Netherlands than in any other country: 19 limit values for recycling and 22 for landfilling. Wallonia also has 19 limit values for recycling, but these include less critical elements such as Al and K. Germany (10-13 limit values for recycling, 11-14 for landfilling), Flanders (8 limit values for recycling, 14 for landfilling) and the EU (15 limit values for landfilling) have regulated significantly less components. France has the least limit values of the regions and countries considered, with 6 for recycling and 9 for landfilling.

It is difficult to compare limit values between countries that use different types of leaching tests. While equilibrium is assumed to have been achieved in agitated extraction tests, this assumption is much less appropriate for column tests. Within one type of leaching test, limit values and leaching results are comparable as long as the L/S ratios are equal. The following comparisons can be made, based on the limit values in Tables 2,3 and on the practical evaluation of two incinerator residues:

- Recycling of bottom ash has to be tested with the column test ($L/S = 10$) both in Wallonia and The Netherlands. The limit values in Wallonia are stricter than in The Netherlands although slightly more heavy metals are regulated in the latter. In The Netherlands extra isolation measures are needed, which are not required in Wallonia.
- According to the limit values, the investigated bottom ash can be recycled in The Netherlands (up to 2006) and France. In Flanders, Wallonia and Germany, the tested bottom ash fraction needs further treatment before recycling. The difference between The Netherlands and Flanders is in agreement with the comparison of limit values.
- For landfilling of non-hazardous waste, the Flemish limit values for category-1 landfills can be compared with the Z_3 and Z_4 landfills in Germany and with the values (EN extraction test, $L/S = 10$) for landfilling non-hazardous waste in the EU. The Flemish limit values are less stringent than the ones for Z_3 and even Z_4 in Germany, although more salts are regulated in Flanders. The EU legislation is stricter than the Flemish one, with respect to limit values, and comparable with the Z_4 category in Germany, although more components (both heavy metals and salts) are regulated in the EU.
- The values for the Category-1 landfill in Flanders are equal to the values for the landfill for hazardous waste in Germany (both DIN extraction test, $L/S = 10$), except for salts where Germany has more components regulated. The limit values for hazardous waste set forth in the European legislation are less stringent than the ones in Flanders and Germany, although more heavy metals are regulated in the EU.

With respect to the investigated fly ash, it could be disposed of in The Netherlands in a C_2 landfill or in a special compartment of a C_3 landfill and in Germany in salt mines. In Flanders, Germany and following the EU legislation, 20% of cement must be added to the fly ash in a solidification/stabilisation process in order to make it acceptable in a landfill for hazardous waste. In this particular example, the EU is as stringent as Flanders and Germany. In France solidification/stabilisation with up to 40% cement would not suffice to make the waste acceptable in a landfill for hazardous waste; in this case another pretreatment is necessary.

The differences in legislation between the European countries may lead to both legal and illegal transport of important quantities of waste between European countries. In order to prevent illegal activities with possibly catastrophical consequences for the environment and human health, harmonization of legislation is needed. To date it is mainly the landfilling legislation that is moving towards harmonization. European legislation on recycling should also be developed soon as the market for recycling can be rapidly increasing nationally as well as internationally.

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Appendix A

The consulted legislation is listed hereafter for each country or region.

A.1. Flanders

- DECREET VAN 2 JULI 1981 (GEWIJZIGD IN 1994) BETREFFENDE DE VOORKOMING EN HET BEHEER VAN AFVALSTOFFEN
- BESLUIT VAN DE VLAAMSE REGERING VAN 1 JUNI 1995 (LAATST GEWIJZIGD IN 2002) HOUDENDE ALGEMENE EN SECTORALE BEPALINGEN INZAKE MILIEUHYGIËNE (VLAREM II)
- VLAAMS REGLEMENT VAN 17 DECEMBER 1997 (VOLLEDIG HERZIEN IN 2004) INZAKE AFVALVOORKOMING EN –BEHEER VAN 1998 (VLAREA)

A.2. Wallonia

- DÉCRET DU 27 JUIN 1996 RELATIF AUX DÉCHETS (DERNIÈRE MODIFICATION LE 16 OCTOBRE 2003)
- ARRÊTÉ DU GOUVERNEMENT WALLON DU 10 JUILLET 1997 ÉTABLISSANT UN CATALOGUE DES DÉCHETS (MODIFIÉ LE 24 JANVIER 2002)
- ARRÊTÉ DU GOUVERNEMENT WALLON DU 14 JUIN 2001 FAVORISANT LA VALORISATION DE CERTAINS DÉCHETS (MODIFIÉ LE 27 MAI 2004)
- ARRÊTÉ DU GOUVERNEMENT WALLON DU 4 JUILLET 2002 ARRÊTANT LA LISTE DES PROJETS SOUMIS À LÉTUDE DINCIDENCES ET DES INSTALLATIONS ET ACTIVITÉS CLASSÉES
- ARRÊTÉ DU GOUVERNEMENT WALLON DU 27 FÉVRIER 2003 FIXANT LES CONDITIONS SECTORIELLES DEXPLOITATION DES CENTRES DENFOUISSEMENT TECHNIQUE (MODIFIÉ LE 18 MARS 2004)

A.3. The Netherlands

- BOUWSTOFFENBESLUIT BODEM- EN OPPERVLAKTEWATEREN-BESCHERMING VAN 23 NOVEMBER 1995
- UITVOERINGSREGELING BOUWSTOFFENBESLUIT VAN 30 JANUARI 1998 (GEWIJZIGD OP 5 OKTOBER 2000)
- STORTBESLUIT BODEMBESCHERMING VAN 20 JANUARI 1993 (GEWIJZIGD OP 5 JANUARI 1998)
- BESLUIT STORTPLAATSEN EN STORTVERBODEN AFVALSTOFFEN VAN 8 DECEMBER 1997 (LAATST GEWIJZIGD OP 13 JULI 2001)

A.4. Germany

- ABFALLGESETZ
- GESETZ ZUR FÖRDERUNG DER KREISLAUFWIRTSCHAFT UND SICHERUNG DER UMWELTVERTRÄGLICHEN BESEITIGUNG VON ABFÄLLEN
- VERORDNUNG ZUR BESTIMMUNG VON ÜBERWACHUNGSBEDÜRFTIGEN ABFÄLLEN ZUR VERWERTUNG
- TA-SIEDLUNGSABFALL
- TA-ABFALL
- LAGA MITTEILUNGEN, ANFORDERUNGEN AN DIE STOFFLICHE VERWERTUNG VON MINERALISCHEN RESTSTOFFEN/ABFÄLLEN – TECHNISCHE REGELN

A.5. France

- ARRÊTÉ DU 18 DÉCEMBRE 1992 RÉLATIF AU STOCKAGE DE CERTAINS DÉCHETS INDUSTRIELS ULTIMES ET STABILISÉS POUR LES INSTALLATIONS EXISTANTES
- ARRÊTÉ DU 18 DÉCEMBRE 1992 RÉLATIF AU STOCKAGE DE CERTAINS DÉCHETS INDUSTRIELS ULTIMES ET STABILISÉS POUR LES INSTALLATIONS NOUVELLES
- ARRÊTÉ DU 9 SEPTEMBRE 1997 RÉLATIF AUX DÉCHARGES EXISTANTES ET AUX NOUVELLES INSTALLATIONS DE STOCKAGE DES DÉCHETS MÉNAGERS ET ASSIMILÉS.

- CIRCULAIRE DU 9 MAI 1994 RÉLATIVE À LÉLIMINATION DES MÂCHEFERS DINCINÉRATION DES RÉSIDUES URBAINS

A.6. The European Union

- DIRECTIVE 1999/31/EC OF 26 APRIL 1999 ON THE LANDFILL OF WASTE
- COUNCIL DECISION 2003/33/EC OF 19 DECEMBER 2002 ESTABLISHING CRITERIA AND PROCEDURES FOR THE ACCEPTANCE OF WASTE AT LANDFILLS

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Assessing the impact of land-applied biosolids from a thermomechanical (TMP) pulp mill to a suite of terrestrial and aquatic bioassay organisms under laboratory conditions

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Abstract

The potential impact on a variety of bioassay organisms when pulp-mill biosolids from a thermomechanical pulp mill (western Canada) were applied to a reference soil has been investigated in a laboratory setup. The current research assessed acute, chronic, and reproductive impacts using a battery of terrestrial and aquatic organisms. Terrestrial organisms were exposed to soil amended with different concentrations of biosolids, while aquatic organisms were used to assess the impact of biosolids' runoff into receiving waters. The former bioassays showed that an application rate of 20 tonnes hectare^{-1} (t ha^{-1}) "bone-dry" biosolids applied to reference soil produced no observable adverse impact on the terrestrial organisms. In the latter assays, undiluted (100%) and 50% diluted biosolids' runoff into receiving water had a detrimental impact on the aquatic organisms. However, concentrations not exceeding 25% (environmentally relevant concentrations) had neither an acute nor chronic impact compared to reference populations. The organisms' abilities to reproduce were also unaltered. While this study only examined the biosolids from one mill, there is the potential that land-application of characteristically well-defined pulp mill biosolids *may* constitute an acceptable way of disposing of pulp and paper mill biosolid residues. However, the biosolids coming from different mills, with differing processes, must be dealt with on a case-by-case situation. Each series of biosolids must be rigorously tested for toxicological impact in the laboratory under tightly controlled conditions. Subsequently, field experimentation must be conducted before definitive conclusions can be made.
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1. Introduction

In recent years, a movement towards minimizing pulp mill discharges into surface receiving waters resulted in an increase in biosolids and sludge produced from the pulping process. Disposal of these solid wastes include landfilling or incineration, but increasingly high costs have led to an evaluation of viable and cost-effective alternatives. The Canadian pulp and paper industry generates 1.7 million dry tonnes of mill biosolids every year (OFIA, 1996;Reid, 1997) and the land-application of this material to forest soil is considered a highly effective alternative to land-filling (Henry and Cole, 1997;Bates, 1999). Organic waste components are potentially degraded by indigenous microbial populations and nutrient constituents are utilized by the region's vegetation (Kays et al., 1997). Many studies have been conducted attesting to the benefits of land-application of sludges (Henry et al., 1993;McDonald et al., 1994;Beyer et al., 1997;Camberato et al., 1997;Macyk, 1999; Tripepi et al., 1994) indicating increased tree growth, increased crop nutrition, and improved soil physical conditions. However, while the high nutrient and carbon content of the biosolids might offer significant benefits to soil

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productivity and tree growth, biosolids application systems which exceed the retention and assimilation capacity of the soil and associated vegetation may have impacts on receiving waters and vegetative productivity. Research conducted by Stanosz et al. (1995) suggest that forest productivity can be threatened where sludge is applied, as a result of nutritionally intensified effects of moisture stress. Also, while soil has a very limited function as a transport medium, it can be an important source for contamination of groundwater should sludge constituents prove to have polluting properties (Lokke and Van Gestel, 1998).

Pulp mill biosolids are usually a mixture produced in the primary clarifiers and then sedimentation of particles during the biological treatment of the wastewater in secondary treatment (NCASI, 1993). The solids content of primary clarifier material is approximately 20–45% and on a dry weight basis, these consist of approximately 85% wood fibres, 10% calcium carbonate, and 5% inorganic clay and ash particles. The nutrient content of the primary clarifier biosolids is low, with a high C:N ratio. Conversely, secondary treatment biosolids have a high organic content that is composed primarily of microbial biomass. These wastestreams are supplemented with nitrogen and phosphorus which are retained by the biomass and can become major components of the secondary biosolids (NCASI, 1993). As a result of the varying compositions of primary and secondary biosolids, the properties of final biosolids may range anywhere from 5% to 75% secondary treatment biosolids (NCASI, 1993).

Most research on land-application of municipal or pulp and paper biosolids has focussed on nutrient cycling and the ability of vegetative cover or plantation forests to utilize the added nutrients. However, the long-term impact on different groups of organisms of these disposal practices are unknown and there are potential problems which must be addressed if sustainable forest management practices are to continue with the land-application of biosolids. There is a paucity of information on the fate and impact of the biosolid constituents themselves, both to terrestrial organisms native to forest biomes and to aquatic organisms when biosolids' runoff impacts surrounding receiving waters. Surface receiving waters and groundwater quality may also be affected by high nutrient level leachate from land-applied material (Ferrari et al., 1999). Certainly, the capacity of a forest ecosystem to process biosolids without causing adverse environmental impact cannot be predicted until a holistic environmental evaluation of the ecosystem receiving the biosolids has been conducted, involving both rigorously controlled laboratory assessment and field-monitoring of the amended area. The overall objective of this study was to assess the environmental impact of land-applied biosolids originating from the pulp and paper industry using both field and laboratory techniques. While the field results are reported in McCarthy et al. (2004) and will not be reiterated, the laboratory-scale setup and observations will be reported here.

2. Methodology

2.1. Biosolids collection

The biosolids for this study came from a thermomechanical newsprint mill (TMP) in western Canada where hardwood and softwood chips are pressurized into high-quality pulp. Total retention time of effluent is 6 days, with the secondary clarifier being an aeration lagoon. While the biosolids constitute a mixture of primary (70%) and secondary material (30%), the bulk originates from the primary clarifier. Under normal conditions, the biosolids are land-applied within 12 h of being removed from the treatment system. However, in the present study, the biosolids arrived in the laboratory within 48 h of being collected and were stored for up to two weeks at 4 °C prior to being incorporated into bioassays. Upon use, they were aerated for 24 h, thereby liberating any gaseous or volatile compounds that had accumulated during storage, after which time, the organisms were added.

For the entire series of laboratory experiments, commercial topsoil (ASB Greenworld^{TM} brand) consisting of 55% clay and silt, 35% water, and 10% organic material was used as a reference substrate.

2.2. Terrestrial bioassays

Table 1 contains the sources and standard growing conditions for all the organisms tested.

2.2.1. Lumbricus terrestris (earthworm)

The earthworm *L. terrestris* was chosen as the soil invertebrate test organism in this study. Of the phylum Annelida and class Oligochaeta, it is widely distributed in soils across temperate North America. Earthworms have hermaphroditic reproductive systems and the earthworm secretes a cocoon. Typically, only one worm per cocoon develops fully, emerging from the cocoon two to three weeks following fertilization. In the current study, sublethal bioassays examined the number of cocoons and subsequently, the presence of successful emergents as a measure of chronic impact and reproductive effect.

2.2.2. Lumbricus bioassay conditions

A three-months bioassay was established for survival and reproduction. The experimental setup included a reference treatment (0 tonnes hectare^{-1} (t ha^{-1})) and a biosolids treatment (20 t ha^{-1} equivalent dry weight).

Table 1
Test organisms

Organism	Source	Growth medium	Photoperiod (light/dark)	Temperature (°C)	Nutrients
Daphnia magna	Ward's Scientific	Dechlorinated water	(16/8) h	24	YCT
Hyalella azteca					
Gambusia affinis					Tetramin®
Selenastrum capricornutum		AAP[a]			Nutrients in specific media
Lemna minor		GM[b]			
Lumbricus terrestris		Organic top soil	No light	12	Worm Food[c] (1/week)
Brassica rapa	Wisconsin scientific		(16/8) h	24	Dechlorinated water (daily)

Source and growing conditions.
[a] Algal Assay Procedure growth media (Miller et al., 1978).
[b] Growth media (USEPA, 1996).
[c] Super Gro™ by Frabill Inc.

The biosolids were applied to organic soil in 20 L cylindrical plastic containers (30 cm height). Subsequently, six earthworms were added to each container (Elvira et al., 1996) and three replicate systems were set up. Humidity in each container was maintained (65–75%) by sprinkling dechlorinated water on the substrate surface (Berry and Jordan, 2001). Acute toxicity, with mortality as an endpoint, was assessed after 4 days, and subsequently, if survival was observed, the earthworms were examined weekly. Reproductive capability was assessed using cocoon production as an endpoint in each sample after three weeks of incubation. Mortality and cocoon numbers were recorded (Lokke and Van Gestel, 1998).

2.2.3. *Brassica rapa (mustard plant)*

Species in *Brassica* genus belong to the mustard family or Brassicaceae and have an average lifecycle of 45 days. These plants are dicotyledonous angiosperms. Plants flower approximately 14 days after planting, and upon artificial pollination, seed pods start to develop. By day 40, seeds can be harvested, and, if the research warrants such testing, a new lifecycle assessment can begin.

2.2.4. *Brassica bioassay conditions*

The test chosen was a 45-day lifecycle test. The equivalent of 0 $t\,ha^{-1}$ (reference), 20, 100 and 200 $t\,ha^{-1}$ biosolids were variously added to the top of 1.5 kg reference soil. The rates were derived from industry and environmental agencies' guidelines and are "typical" according to the USEPA (1997) (20 tonne equivalent dry weight ha^{-1} year) and MOE/OMAFRA (1996) (10–40 tonne equivalent dry weight ha^{-1} year). Industry trials (Alberta Research Council, 2001) used 10–80 $t\,ha^{-1}$. The test was performed in three replicate trays (33.5 cm-L × 20.5 cm-W × 9.0 cm-H) and in every tray, 15 *Brassica* seeds were planted (three rows of five holes of 0.5 cm depth). The trays were placed under light banks and watered daily. After flowering, cross-pollination was performed manually using sterile cotton swabs. The number of successful germinations was assessed, along with the number of flower and seedpods that developed. Due to the branched nature of the roots, an average root length was not possible. Thus, an average maximum root length of each germinated plant was measured for each replicate tray, alongside a minimum root length. This ratio was recorded as the root-length index.

2.3. *Aquatic bioassays*

2.3.1. *Run-off setup*

The potential in the natural environment for rain to wash land-applied biosolids into adjacent receiving waters and impacting aquatic organisms is great and this scenario was assessed in the laboratory using simulated environmental conditions. In order to obtain environmental runoff conditions, a laboratory "slope-simulator" was designed (Fig. 1). A wooden ramp was built capable of supporting 60 kg of biosolids and soil. The ramp dimensions were 240 cm × 36 cm and was inclined to simulate a 15% slope, which is within the acceptable range of real-world land-applications of biosolids (USEPA, 1997). The ramp was lined with new food-grade plastic foil for each runoff trial. For the biosolids-amended soil simulation, 1.5 kg of biosolids were spread over the surface of the reference soil on the ramp according to a 20 $t\,ha^{-1}$ application rate. This value is consistent with application rates identified in pilot studies examining the use of pulp and paper biosolids as forest soil fertilizers (industry report – source confidential). For rainfall simulation, 16 L of deionized water were sprinkled over the surface of the substrate, corresponding to an average one-day rain event of 20 mm. At the lower end of the ramp, a large fibreglass mesh (1 mm) was installed to filter the coarse material. The water runoff, together with the finer material washed away from the surface soil, was collected in a glass receiving-vessel. This mixture of water and fine particles was used in the different aquatic bioassays.

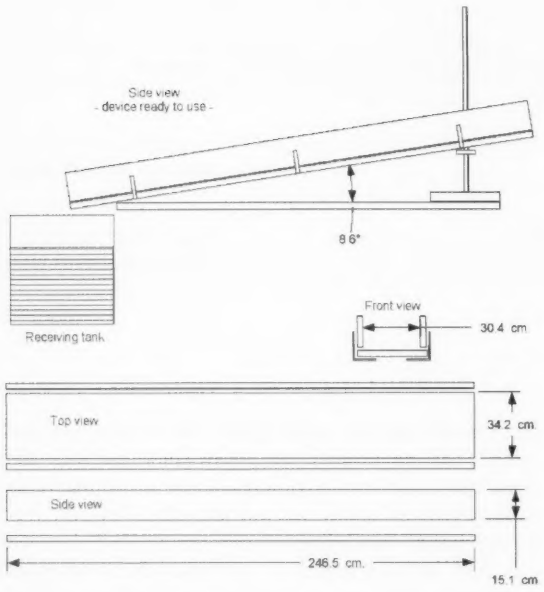

Fig. 1. Laboratory simulation tray for collecting water runoff originating from soil surface.

2.3.2. Runoff samples preparation

Runoff from biosolids-amended soil and from reference soil were tested in parallel. For all of the aquatic bioassays, a series of diluted runoff samples were prepared. The dilutions, using dechlorinated tap water, were 0% (blank), 10%, 25%, 50%, and 100% (v/v). Water quality (pH, temperature, hardness, and alkalinity) was monitored on a regular basis throughout the exposure period. During chronic exposure static tests, deionized water was added every second day to compensate for evaporation.

2.3.3. Daphnia magna (waterflea)

The impact of constituents desorbed from the runoff biosolids and partitioned into the overlying water was assessed using the watercolumn zooplankter *D. magna*. These organisms have been extensively studied in toxicological research and a large database exists documenting the appropriateness of their use. The bioassays conducted in the current study included acute toxicity bioassays and chronic lifecycle tests. While lethal endpoints were obvious indicators of toxicity, sublethal responses such as reproductive potential and numbers of neonates were also used to assess impact.

2.3.3.1. Daphnia bioassay conditions. Organisms were cultured in the Ryerson laboratory according to protocol established by the USEPA (1993). A 21-day static renewal bioassay was initiated that assessed survival and reproductive performance of the invertebrates, according to protocols of Environment Canada (1990) and USEPA (1993). Initially, the runoff water was centrifuged for 10 min at 3000 rpm to reduce the level of suspended solids because high concentrations of particulate matter can clog the filtering mechanism of these invertebrates. Subsequently, successive dilutions of runoff, 10 replicates each, were prepared in 50 mL beakers. Acute mortality was recorded after 24 h, and where mortality was not observed, the animals were subsequently assessed for survival and number of offspring for each sample concentration. This was conducted every second day for the rest of the experiment (21 days in total). Toxicity results were calculated as a percentage of the numbers of *Daphnia* in reference samples, where survival rates exceeded 80%, as stipulated by Nebeker et al. (1984).

2.3.4. Hyalella azteca (benthic amphipod)

H. azteca (Class: Crustacea, Order: Amphipoda) are omnivorous benthic crustaceans found in shallow freshwater lakes throughout North America and were highly recommended as sediment test organisms by de March (1981) and Nebeker et al. (1984). They are a major component of the foodweb in ecosystems such as the Great Lakes (Winell and Jude, 1987; Borgmann and Munawar, 1989), have short lifecycles (two to four months), and survival, growth, and reproduction can be monitored in 21-day partial lifecycle toxicity tests (Reynoldson and Day, 1993). Thus, they were considered an excellent bioassay organism in the current study to assess the toxic impact of biosolids present in the receiving water due to runoff events and were cultured at the Ryerson laboratory according to Borgmann and Munawar (1989).

2.3.4.1. Hyalella bioassay conditions. A 21-day static-renewal bioassay to assess acute, chronic, and reproductive impairment was conducted using protocols developed by Borgmann and Munawar (1989) and the USEPA (1994). Successive dilutions, five replicates each, of runoff were prepared in 500 mL beakers filled with 200 mL solution. The suspended particulates in all solutions were allowed to settle for 24 h prior to the addition of the amphipods. Three neonate amphipods were introduced to each beaker. Twenty-four hours later, surviving organisms in all test chambers were counted to assess potential acute toxicity. If acute toxicity was not apparent, the bioassay continued for 21 days, whereupon the young and surviving organisms were counted to determine the effect of effluents on the survival, growth, and fecundity.

2.3.5. Selenastrum capricornutum (green algae)

S. capricornutum is a non-motile, unicellular crescent-shaped green alga (Class: Chlorophyceae) whose normal

uniform morphology allows for ease of identification and enumeration. Phytoplankton populations are considered essential in toxicity testing because of their importance in the aquatic ecosystem as primary producers. Phytoplankton are easy to culture, have short generation times, and were found to be sensitive to many contaminant compounds (Giesy and Hoke, 1989).

2.3.5.1. Selenastrum bioassay conditions. A 72 h chronic toxicity bioassay, adopted from Blaise and Couture (1984), was established on microplates. Runoff samples were centrifuged for 30 min at 5000 rpm and subsequently filtered through 0.45 μm ultrafine, glass microfibre Whatman® GF/Cfilters. Successive dilutions of runoff, four replicates each, were added to 2 mL wells on 4 × 6 well Corning® microplates, along with an initial concentration of 10^5 algae cells mL^{-1}. The samples were incubated for 72 h at 24 °C and 4000 lux (Miller et al., 1978). The number of cells ml^{-1} was counted optically using a microscope and a Fuchs-Rosenthaal haemocytometer (Singh, 1973).

2.3.6. Lemna minor (duckweed)

The macrophytic angiosperm *L. minor* (Family: Lemnaceae) is a monocot inhabiting still or slow-moving freshwaters distributed throughout the world. Due to its free-floating existence, it derives most of its nutrients from the watercolumn. It has a rapid reproductive rate; the doubling time in laboratory cultures has ranged from 0.35 to 2.8 days (Hughes et al., 1988).

2.3.6.1. Lemna bioassay conditions. A 21-day chronic toxicity bioassay (USEPA, 1996) was established. Successive dilutions of runoff, four replicates each, were distributed to 2 mL wells in 4 × 6 well Corning® sterile microplates. Each well was spiked with a nutrient-rich medium (reference medium) and one plant with four fronds was added. The samples were incubated for 21 days at 24 °C and 4000 lux. The number of fronds present in each well after the incubation period was recorded (USEPA, 1996).

2.3.7. Gambusia affinis (mosquitofish)

Fish have been used extensively in monitoring bodies of water for toxicity impact and are considered as sensitive as invertebrates to many contaminants (Reynoldson and Day, 1993). The mosquitofish belong to a family of live-bearing tooth carps, the Poecilidae, which are now native to North America, having been introduced two centuries ago from Europe. They are of practical value in the current study due to their small size (3–4 cm), ease of culturing, and insensitivity to changes in the physicochemical parameters of water.

2.3.7.1. Gambusia bioassay conditions. A 96-h acute toxicity test was established in the current study, with successive dilutions of runoff, three replicates each, prepared in 10 L glass tanks. One fish per tank was introduced, and the test was performed on three replicates of each dilution. Mortality of the fish was checked daily.

2.4. Statistical analysis

The data from this study represent proportions of the specific populations exposed to different ratios of land-applied biosolids or from differing concentrations of runoff originating from biosolid-amended soil. The statistical method for testing the differences in response from the various organisms to those treatments is the Chi-square (χ^2) analysis non-parametric method (Hoshmand, 1997). This test offers a powerful and robust detection of differences among populations of discrete proportions for which normal distribution and parametric tests do not apply. The level of significance considered for the study was 0.05. The test allows us to state if certain concentrations of biosolids or biosolid runoff had a significant impact on all populations tested.

For the *Daphnia* and *Hyalella* reproduction bioassays, the mean number of neonates per live organism was calculated for each concentration of runoff tested. The *t*-distribution was used as a statistical method to make inferences about the different neonate means obtained from the invertebrates exposed to various concentrations of runoff. This method applies to small samples from populations approximately normally distributed with equal population standard deviations (Hoshmand, 1997). The *t*-test was performed at the 0.05 level of significance and determined if any runoff concentrations had an impact on the mean number of neonates when compared to the reference treatment.

3. Results

3.1. Terrestrial bioassays

Qualitative observations for the *Lumbriculus terrestris* bioassays indicate that all earthworms appeared healthy with intact, moist cuticles. Quantitative results obtained with the earthworms are presented in Fig. 2. Survivorship averaged four animals out of the initial five earthworms placed in the reference soil and 4.6 animals in biosolids-amended soil among the three replicates for each treatment. The average number of cocoons found in the two soils (reference and biosolids-amended) was 11.5 and 10.5 respectively. In

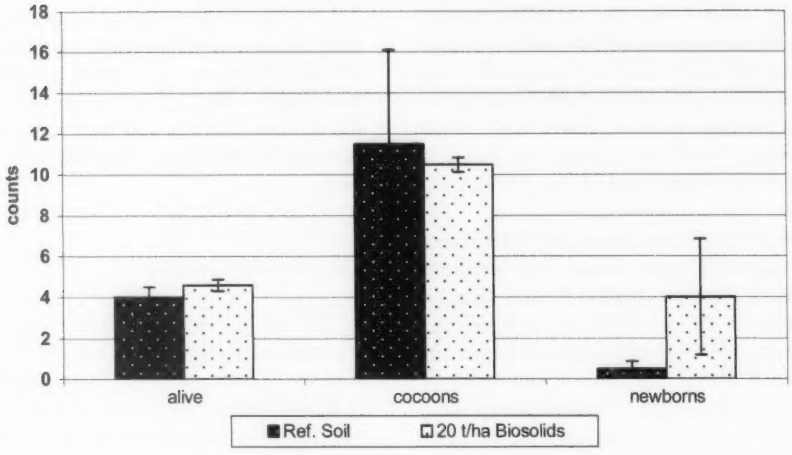

Fig. 2. Earthworm response to 20 $t\,ha^{-1}$ biosolids added to culture soil.

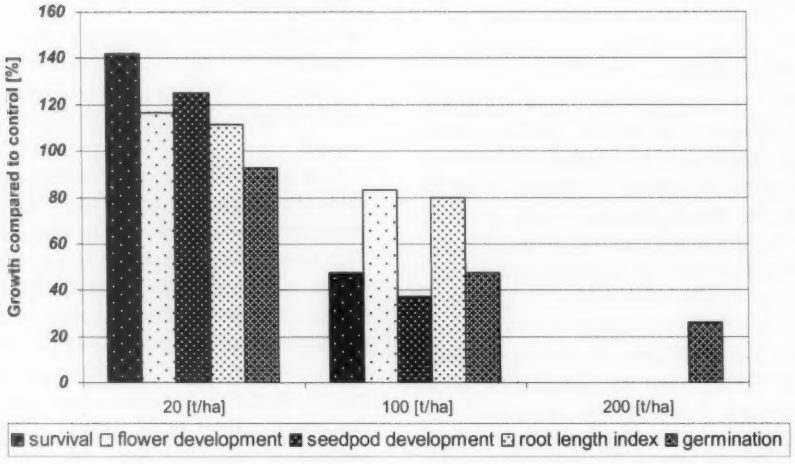

Fig. 3. Different parameters assessing *B. rapa* development in biosolid amended soil.

both soils, juvenile earthworms could also be found (Fig. 2).

In the *B. rapa* phytoexperiments (Fig. 3), an application rate of 20 $t\,ha^{-1}$ biosolids lead to enhanced plant survival (142%), flower and seedpod development (116% and 125% respectively), and a root-length index of 111% compared to plants grown in reference soil. Only the germination rate decreased by 8–92%. However, rates of 100 and 200 $t\,ha^{-1}$ of biosolids had an adverse effect on these various endpoints. Germination was substantially reduced to 47% for the 100 $t\,ha^{-1}$ biosolids application rate and to 26% for 200 $t\,ha^{-1}$ biosolids addition. At 100 $t\,ha^{-1}$, plants that germinated did develop flowers, seedpods, and their root-length index averaged 79% compared to the plants grown in reference soil. However, in the 200 $t\,ha^{-1}$ treatment, the plants did not survive to complete their life cycle (Fig. 3).

3.2. Aquatic bioassays

The aquatic organisms were exposed to a series of dilutions from the biosolids' runoff. Immediate toxic impact (Fig. 4) was marginal when the concentration of the runoff did not exceed 25%. At these low concentrations, *D. magna* showed no mortality after 24 h of exposure. The exposure to 50% dilution runoff lead to the death of half of the *D. magna* population and 100% runoff was acutely toxic to the entire population. *H. azteca*, after 24 h exposure to 10%, 25%, and 50% biosolids-amended runoff had very high survival rates of 100%, 87%, and 80% respectively. However, undiluted biosol-

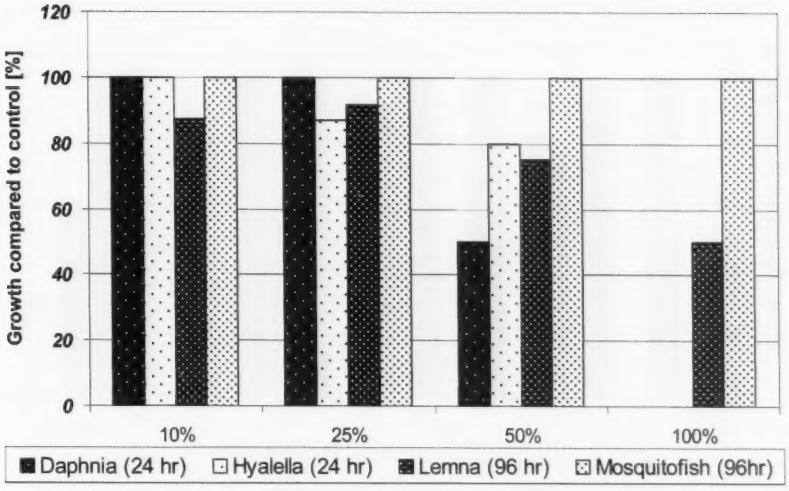

Fig. 4. Acute bioassays on different concentrations of runoff from biosolid amended soil.

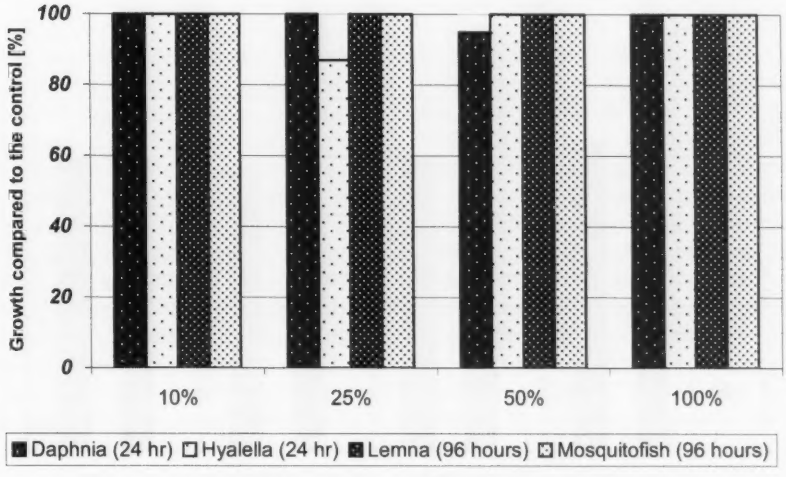

Fig. 5. Acute bioassays on runoff originating from reference soil.

ids-amended runoff killed the entire population. While the development of *L. minor* frond growth showed similar values at 10%, 25%, and 50% concentrations when compared to the reference culture medium, the 100% runoff allowed the development of only 50% of fronds, compared to the culture medium. No acute mortality could be seen with the mosquitofish *G. affinis* upon 96 h exposure to all concentrations of biosolids-amended runoff (Fig. 4).

The runoff from reference soil showed no inhibition of the endpoints of the various organisms (Fig. 5).

Chronic toxicity bioassays from the biosolids-amended runoff on the aquatic plants and invertebrates are shown in Fig. 6. Results indicate that at 10% and 25% runoff concentrations, *D. magna* survival rate continued to be 100% after 10 days of exposure. However, deleterious impact was noted at 50% concentrations, where only 14% of the daphnids continued to survive. *H. azteca* was exposed to different concentrations of runoff for 21 days. Fig. 6 shows an increasingly negative effect of the biosolids-amended runoff, with only 67% and 53% survival rates for 25% and 50% runoff concentrations respectively. Since *Daphnia* and *Hyalella* had total mortality in 100% runoff during acute bioassays, chronic toxicity tests were not possible.

After 19 days exposure to biosolids-amended runoff concentrations of 10%, 25%, and 50%, *L. minor* had frond development consistent with growth found in the artificial growth medium (reference treatment). It was only during exposures to 100% runoff concentration that impact was noted, with frond development

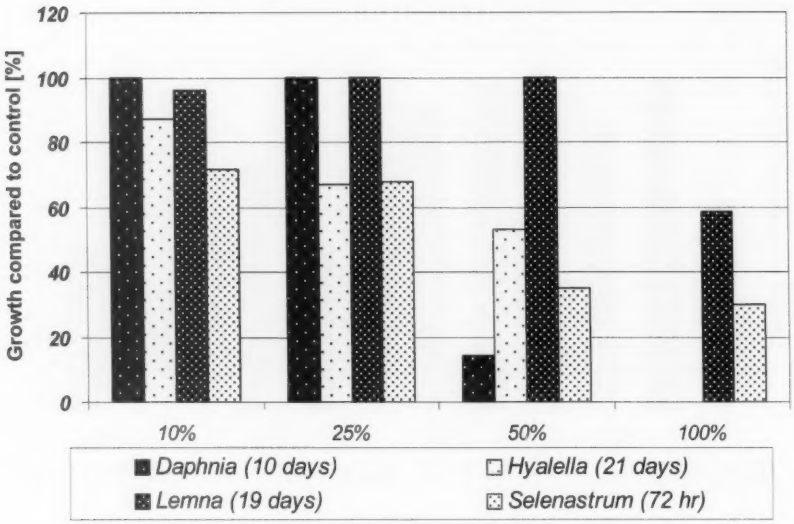

Fig. 6. Chronic bioassays on different concentrations of runoff from biosolid amended soil.

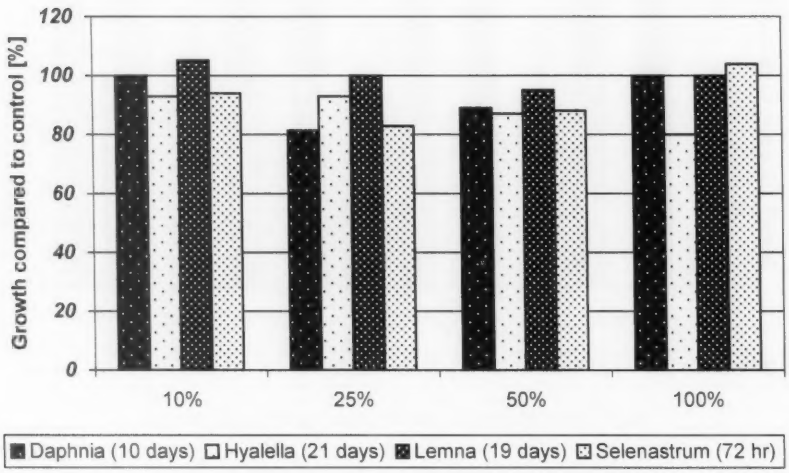

Fig. 7. Chronic bioassays on runoff originating from reference soil.

reduced to 58% of the numbers obtained in the growth medium. Conversely, in chronic bioassays using the green algae *S. capricornutum*, diminished growth was observed at levels as low as 10% concentrations and continued to decrease with increasing concentrations of biosolids-amended runoff when compared to their growth in artificial culture medium.

When runoff originating from the reference soil was tested for chronic toxicity, survivorship was over 80% regardless of the organism tested (Fig. 7).

For assessing the reproductive impact on the aquatic invertebrates, the average number of offspring produced by live adults is presented in Fig. 8. The number of *D. magna* offspring averaged between 26 and 33 neonates per live adult when runoff concentration ranged from 0% to 50%. For *H. azteca*, in the blank sample (0% runoff) and in 10% runoff, the average values of neonates per organism were of 13 and 15 respectively. Concentrations of 25% and 50% runoff induced a decrease in the number of neonates per live adult, with values as low as 2 and 3 neonates per *H. azteca* organism respectively. Since *D. magna* and *H. azteca* had 100% mortality during acute toxicity bioassays (100% concentrations), no offspring were recorded.

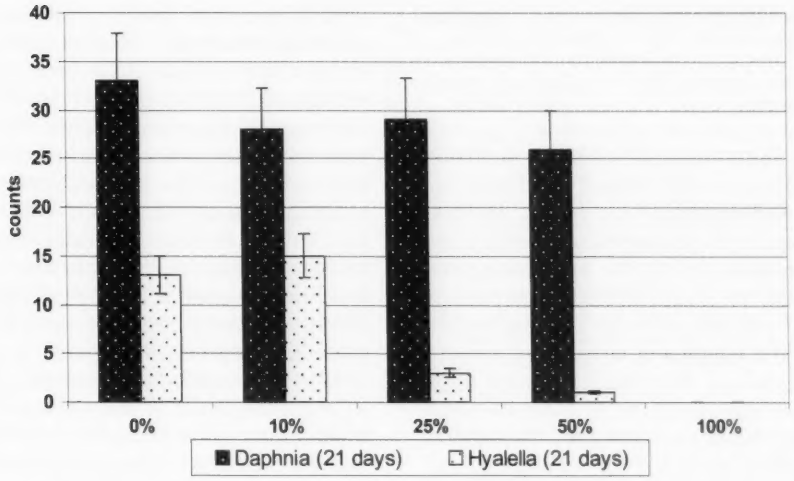

Fig. 8. Average number of offspring produced by live adult in *Daphnia* and *Hyalella* reproduction assays on runoff from biosolid amended soil.

4. Discussion

Previous studies in our laboratory examined potential impacts on *L. terrestris* and the lettuce *Lactuca sativa* from land-application of municipal sewage treatment plant sludge (McCarthy et al., 1999). Decreased survivorship in earthworms and an impaired germination rate in the plants were observed. In the current study, the laboratory experiments were meant to evaluate the potential biological impact of land-applying different concentrations of pulp mill biosolids to reference soil. While changes in the geochemistry of major cations such as Ca^{2+}, Mg^{2+}, Na^{+} and SO_4^{2-} were previously investigated and results showed little impact during a one-time application of 40 $t\,ha^{-1}$ biosolids in a forested watershed (Kraske and Fernandez, 1993), the potential detrimental impact of land application of biosolids to forest biocenosis had, until now, received little attention (McCarthy et al., 1999). The focus of the current study, therefore, was to develop a holistic approach, using both terrestrial and aquatic organisms, to assess impact under rigorously controlled laboratory conditions.

4.1. Terrestrial bioassays

4.1.1. Brassica rapa

Using χ^2 statistical analysis, results from *B. rapa* (reference soil versus different ratios of biosolids (20, 100 and 200 $t\,ha^{-1}$)) indicated that certain ratios did affect the physiological integrity of this species. However, when the results of the different endpoints (germination rate, root elongation, number of flowers, seedpods, and seeds) measured for plants grown in 20 $t\,ha^{-1}$ amended biosolids were compared (χ^2) to plants growing in reference soil, there were no significant differences. This indicated that the detrimental impact is associated with the higher ratios of biosolids (100 and 200 $t\,ha^{-1}$). The slightly higher values of tested endpoints obtained in 20 $t\,ha^{-1}$ amended soil versus the reference soil are statistically irrelevant at the level of significance ($\alpha = 0.05$) imposed on this study. However, it is important to note here that a forest soil is a naturally nutrient-poor soil compared to the organic nutrient-rich soil used as a reference soil in the current study. Since a worst-case scenario was assessed, the impact of biosolid amendment is compared to an optimum theoretical growth condition. This study was designed to assess potential *detrimental* impacts of biosolids application and *not* to redo the experiments reported by Macyk (1999), Montigny and Stearns-Smith (2001), and Morris and Nutter (1996). Those studies showed the beneficial impact upon tree growth of such practices in natural environments with nutrient-deficient forest soils and the validity of those experiments is not contested by the present work. The negative impact noted with higher ratios of biosolids (100 and 200 $t\,ha^{-1}$) on the relatively fragile *B. rapa* may be due to mechanical compaction of the biosolids leading to an impediment of plant growth.

4.1.2. Lumbricus terrestris

The results obtained from earthworms growing in biosolids-amended soil compared (χ^2) to those in reference soil show that land-application of biosolids at 20 $t\,ha^{-1}$ dry material to forest soils does not have a detrimental impact on *Lumbriculus*' ability to survive long-term exposure to biosolids, nor was reproductive ability impaired. Also, the presence of juveniles could be recorded in both reference and biosolids-amended soil.

Higher ratios (100 and 200 $t\,ha^{-1}$), which had already proved inappropriate due to the plant inhibition shown with *B. rapa*, were not tested because these application

rates were considered irrelevant for practical applications.

4.2. Aquatic bioassays

In assessing the impact on aquatic biota from biosolids' runoff into adjacent water bodies, the design and operation of a "runoff simulator" was crucial. The runoff water was collected on the steepest acceptable slope for biosolids amendment (15%). The application ratio of 20 $t\,ha^{-1}$ biosolids was considered sufficient because it is both consistent with the previously cited application rates and it ensured a continuous coverage of about 5 cm of the experimental soil. The amount of water simulating precipitations was calculated in order to match environmentally significant levels (20 mm). The laboratory "runoff simulator" proved to be appropriate and effective for the purpose of the present study and the required quantities of runoff samples were easily collected in a repeatable manner. Future experiments may consider reducing the slope in order to simulate more environmentally relevant terrestrial topographies where biosolids are generally applied.

While assessing the impact of biosolids runoff, a range of concentrations was used, including ratios which were higher than environmentally relevant levels. Concentrations as high as 50% and 100% were used, even though runoff into receiving waters in the environment rarely attains concentrations above 15%. Such high quantities of runoff are associated with torrential rains, well above the 20 mm precipitation simulated by the experiment. At such elevated values of precipitation, the dilution factor of the material carried by the runoff will be highly increased.

By grouping the aquatic organisms together for each runoff concentration (Figs. 4–7), potential impact on the aquatic biota could be established holistically. Although synergism between different groups of organisms could not be tested due to the individual setup of organisms, the potential impact on the biocenosis (Clement and Cadier, 1998) was possible due to the statistical analyses that were conducted. χ^2 statistical analyses were applied to all acute survival rates obtained from all aquatic experiments, including all runoff concentrations (0%, 10%, 25%, 50% and 100%) originating from biosolids-amended soil and reference soil. The test indicated that certain runoff concentrations caused acute impact on the bioassay organisms. However, the null hypothesis was accepted (indicating that there is no statistical difference between control and treatments) if the same statistical analyses were applied only to data obtained from organisms exposed to environmentally relevant concentrations (0%, 10%, and 25% runoff). The interpretation of this statistical result suggests that runoff from areas where biosolids were land-applied at a rate of 20 $t\,ha^{-1}$ does not have an acute impact on aquatic biota when concentrations of runoff water are below 25% in a receiving waterbody. This interpretation is based on the results from a suite of different groups of organisms representing different levels of the aquatic foodweb (primary producers including phytoplankton and macrophytes, invertebrates, and vertebrates) living in different parts of an aquatic biotope, acutely exposed to diluted runoff. This result is further confirmed by the fact that there are no statistically significant differences (according to χ^2 test) between runoff originating from reference soil at any tested dilution (from 0% to 100%) and results from 10% and 25% runoff from biosolids-amended soil.

Identical overall observations can be made from the χ^2 testing of results of chronic toxicity, indicating that while runoff concentrations from biosolids-amended soils do not exceed 25% into a receiving waterbody, no potential detrimental impact on aquatic biota could be observed.

Concurrently, the reproductive capacity of *Daphnia* and *Hyalella* was assessed. A two-sample *t*-test of means was performed. This was possible because the average number of neonates had been statistically asserted as being a small sample of a normal distribution population. Each concentration (except 100% runoff which was acutely toxic so no neonates were produced) was compared to results obtained in the reference cultures. For the biosolids-amended soil, regardless of the concentration of biosolids runoff, the reproductive capacity of *D. magna* was not affected. Similar analysis performed on *H. azteca* indicated statistically significant decreases in neonate production when adults were exposed to 25% and 50% runoff. The 10% concentration of runoff did not appear to impact the amphipods' capacity to reproduce.

5. Conclusion

To assess the potential deleterious impact of land-applied pulp-mill biosolids both to the terrestrial ecosystem and to the adjacent receiving waters, a laboratory strategy was developed that involved a myriad of organisms representing terrestrial and aquatic biota.

The testing of terrestrial organisms exposed to different application ratios of the biosolid showed that if these ratios do not exceed 20 $t\,ha^{-1}$ equivalent of bone dry weight, no adverse impact on plants or on earthworms were observed either acutely or chronically.

Whenever biosolids runoff did not exceed 25% concentration in receiving water, there was no impact on the aquatic organisms compared to biota grown both in artificial culture media and exposed to runoff from reference soil. The organisms were not acutely or chronically impacted, and their ability to reproduce was also unaltered. These findings concurred with those observed

in a field study conducted by McCarthy et al. (2004) where experiments were carried out on experimental forest plots amended with biosolids from the mill used in the current study. The field study indicated that no obvious impact could be found from pulp mill land-application on selected terrestrial species or aquatic organisms from run-off into receiving waters.

From these laboratory experiments, and corroborated by field observations (McCarthy et al., 2004), it may be concluded that in specific situations such as the current one, land-application of pulp-mill biosolids to forest ecosystems may constitute an environmentally sound management of pulping waste residues. However, biosolids from each mill must be tested on a case-by-case situation before being land-applied, initially in the laboratory under rigorously controlled conditions such as the situation outlined in the current study, and subsequently tested under field conditions. The current study ultimately may assist in developing regulatory guidelines and strategies in the assessment of land-applied pulp-mill biosolids.

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Improving the solid waste management in Phnom Penh city: a strategic approach

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Abstract

Though the solid waste management (SWM) system in Phnom Penh city in general has been upgraded since the waste collection service was franchised out to the private sector, the performance of the existing SWM system is still low. Unreliable and irregular collection service still exists. This means that there are shortcomings in the existing SWM system that need correction. This paper is an attempt to identify those shortcomings in order to find ways to improve the existing system. First, the present SWM system is reviewed. Then the system is evaluated to find constraints and shortfalls and finally some appropriate strategies are proposed that may help make SWM in the city more effective and efficient to meet environmentally sound objectives.
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1. Introduction

It is widely recognized that solid waste management is not only a technical problem but it is strongly influenced by political, legal, socio-cultural, environmental, and economic factors, as well as available resources. Moreover, these factors have interrelationships that are usually complex in the waste management system. It is suggested that appropriate solutions to the complex waste management problems be sought from a system perspective, taking into account all of the above factors in the local area (Mashayekhi, 1993; Vesilind et al., 2002). Failing to do this, solutions proposed to solve solid waste issues may not be effective enough to yield any fruitful results. This is true for the case of solid waste problems in Phnom Penh city.

The municipality of Phnom Penh (MPP), which has the responsibility for SWM in the city, lacks financial resources to operate its own solid waste collection and disposal system. With the public pressure to improve the solid waste service in the city, and due to its financial resource constraints, the MPP has franchised out solid waste collection service to a private company. This certainly solves the cash flow problems of the MPP, as the contractor can recover its costs from the collection of service fees from its customers and thus, there are no direct capital contributions from the municipality. More importantly, the strategy can help the MPP to respond to the solid waste service demands of the city dwellers. This seems to be a considerable achievement. However, the overall performance of the contracted SWM service is still low and littering remains very high. This clearly shows that solving the financial resource issue alone does not mean solving solid waste issues. The solid waste problems prove generally to be much more complex. In spite of this, solutions to waste problems must be found to avoid potential threat to the public health and urban environmental quality in general. Hence, it is necessary to evaluate critically the existing system and develop appropriate strategy to improve SWM in the city.

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2. Baseline conditions

2.1. National priority policy

According to the Socio-Economic Development Plan (SEDP) prepared by the Ministry of Planning (MOP, 2002a), the government strategy to meet the objective of the economic growth focuses strongly on agricultural and rural development, and to support economic growth, the government strategy is to allocate resources to restore and expand critical economic and social infrastructure. The strategy is officially documented in the Public Investment Program (PIP) prepared by the MOP (2002b) that is used by international aide agencies, donors, and development banks for selecting and financing appropriate projects.

The resource allocation to public sectors under the PIP has been split into seven sectors: (1) agriculture, (2) energy, (3) industry and trade, (4) water resource and management, water supply and sanitation, (5) education and training, (6) health, and (7) environment and conservation. Although SWM is included in the environment and conservation sector, the attention to the SWM field does not seem to be adequate. The only SWM project under the current PIP is a new sanitary landfill, and other general issues of SWM system such as improving solid waste service, resource recovery, etc., are not included. It seems that there is a lack of a comprehensive SWM strategy for soliciting external funding.

2.2. Demographic indicator

Phnom Penh, the capital city of Cambodia, has a total population of approximately 1 million which consists of about 0.6 million of urban population and 0.4 million of rural population. The average population density in Phnom Penh is about 3448 persons per km^2 (MOP, 2001). The population growth in the city and in the country as a whole is shown in Table 1.

According to Table 1, the population growth rate in Phnom Penh city is higher than the average of the whole country. This shows that there may be a rapid population inflow into the city, perhaps because of job opportunities or other attractive factors. However, more people means more waste, and more waste means more resources needed for waste management. So, the rapid population inflow should be considered in designing a waste management plan.

2.3. Economic indicators

Cambodia for the past 20 years has been classified as one of the poorest countries in the world. Since the election of May 1993, the Royal Government has taken constructive action to change the chaotic macroeconomic setting that has previously existed. There have been some revenue reforms, expenditure control, legislative and regulatory initiatives, a climate more conducive to privatization, and new domestic and foreign private investment. With such reforms, there is a growth trend in the national economy. Table 2 lists some key economic indicators. However, Cambodia is still one of the poorest countries in the world. GNP per capita of the country is less than 280 USD/year. This is not only much lower than an average of Southeast Asia and Pacific Ocean countries (990 USD per capita/year), but is also below the low-income country average (520USD per capita/year) (Mori, 2000). This factor may affect the ability of the public to afford the high standard of solid waste service to be offered.

2.4. Urban waste characteristics

Waste cannot be effectively managed at any level without a good understanding of what it comprises, how much there is, who makes it, what happens to it, and what its effects are (Rushbrook and Finnecy, 1988). For example, without a knowledge of the composition of the waste, it would be difficult to find an appropriate disposal method (e.g., does incineration make sense and should it be introduced into the waste management system?). Similarly, it would be difficult to design a waste collection system if adequate data on the amount of generated waste to be collected is not available. The urban waste characteristics may be conveniently considered under two headings: waste generation, and composition of the generated waste.

Table 1
Population growth

	1986	1993	1995	1998
In Phnom Penh[a]				
Growth rate (%)	11.3	5.4	2.7	–
Population (thousands)	561	810	854	999
In Cambodia[b]				
Growth rate (%)	2.8	3.3	2.5	2.4
Population (thousands)	7762	9653	10,198	11,599

[a] Mori (2000).
[b] MOP (2001).

Table 2
Key economic indicators for Cambodia

	1996	1997	1998	1999	2000	2001
GDP at current prices (billions of riels)	8271	9125	10,795	11,797	12,149	12,724
GDP at current price (millions of dollars)	3131	3042	2841	3088	3149	3234
GDP per capita (dollars)	273	258	234	248	247	247
Growth rate of real GDP (1993 prices)	6.7	0.7	2.9	6.8	5.5	5.3
Budget revenue (percentage of GDP)	9.1	9.7	8.7	11.2	11.7	12.0
Budget expenditure (percentage of GDP)	17.4	13.8	14.4	16.4	17.3	18.4

Source: CDRI (2003).
Riel: 2003 exchange rate is approximate at 4000 riels per 1 $US.

Table 3
Solid waste generation in Phnom Penh, 1995–2000

	1995	1996	1997	1998	1999	2000
Waste volume (m^3)	363,703	381,610	380,098	450,963	511,000	584,000

Source: MOE (2001).

2.4.1. Waste generation

This study covers only management of municipal solid waste, that is, waste generated by households, shops, offices, restaurants, hotels, markets and industries (only normal non-hazardous waste), as well as street sweepings. Industries producing hazardous or chemical waste and those engaged in construction and demolition will be expected to organize their own waste collection services based on government regulations.

As mentioned earlier, the estimate of the quantity of waste generation to be handled is crucially important to design collection service and disposal facilities. A grossly inaccurate recording of the amount of waste could lead to over or under-provision of collection services or disposal facilities. Unfortunately, in a developing country like Cambodia, data related to SWM is scarce. Moreover, information sharing among the authorities concerned with SWM is extremely limited.

Up to now, there has not been any rigorous estimate of the waste generation per capita in the city of Phnom Penh. The value of waste generation per capita, 0.65 kg/day, is an assumed value based on the data available in other countries with similar economic situations (Mori, 2000). However, there is an estimate of annual generated waste based on counting the number of trucks that go to the only existing dumpsite. Such an estimate may have a very limited accuracy because firstly, the collection trucks are not always fully loaded as they are supposed to be. Secondly, the uncollected waste left in the city in many areas is not taken into account, so the estimated generated waste may be much less than the actual amount. The amount of generated waste is shown in Table 3. According to data in Table 3, the waste generation in the city has increased over time. Better planning for SWM should be established to avoid any serious outcome.

Table 4
Characteristics of solid waste in Phnom Penh

Parameter	Value
Composition (% wet wt.)	
Organic waste	65
Inorganic waste	
Paper/cardboard	3.8
Plastics	13.2
Metal	1
Glass	4.9
Rubber, leather, etc.	0.6
Other	11.5
Moisture content (%)	50
Density (kg/m^3)	350–400
Calorific value (kcal/kg)	1000

Source: Inter-Consult (2002).

2.4.2. Composition of the generated waste

The results of a survey of composition of the generated waste made by Inter-Consult (2002) are shown in Table 4. The results show that the plastic content is unusually high, probably because it was wet, which increased the weight. Much of this material, according to the report, is in small pieces, mixed types, wet or dirty, making it difficult to be recycled. The results of the analysis also show that the generated waste in the city is largely organic matter that has potential for composting.

3. Introduction of the existing SWM system

The philosophy of this paper is that a patient's illness will probably be effectively cured if the cause of the disease is clearly identified. Similarly, SWM problems may be effectively solved if the shortfalls and constraints are clearly understood. Therefore, it is important to review

first the existing arrangement of the SWM system in the city to find shortfalls and constraints in the system before appropriate strategies can be proposed.

3.1. Existing technical arrangement system

3.1.1. Waste storage

Generally, no system for storing waste in bins or containers at the source of household waste generation has yet been introduced in Phnom Penh. Restaurants, hotels and shops usually use their own bins and containers for on-site storage of the waste, but a standardized system of bins and containers that could be mechanically loaded into a collection truck does not exist.

The municipality has placed public bins made from half-oil drums along some of the major roads for temporary storage of waste, to be used by the public to put small amounts of waste such as paper or wrappings, in order to avoid littering, and to improve the cleanliness and appearance of these areas. Unfortunately, the places where drums have been put become points where people throw their household waste. This shows that the public awareness is still very low. Moreover, though the storage bins are available at some places, the number of bins is far below the number required. As a result, the waste is always seen spilled over and scattered around the bins.

3.1.2. Waste collection and transportation

Waste collection service is available only in some places such as at residential areas with good standard roads, shopping centers, restaurants, and hotels. It was estimated that more than 20–30% (real figures could be higher) of the population in the city is without regular or adequate collection service and the coverage, efficiency and frequency of collection vary substantially from one area to another or even from one street to another street (Mori, 2000). Moreover, the transportation of waste to the dumpsite has not been properly managed. Wastes such as plastic bags or other light materials are clearly seen flying from the trucks during transport. This also contributes to the litter on streets.

It seems that the collection service in Phnom Penh at present is deteriorating rather than improving. In many areas, the collection frequency has dropped from once a day to once every two or three days. In addition, the collection times are quite variable. Residents must wait for the trucks to honk their horns to signal that they are on the street. However, with irregular collection times, people are not always at home or ready to deliver the waste. Consequently, more and more households place their waste outside their houses along the curb or roadside. The waste bags are subject to being torn by scavenging animals that search for something to eat or by waste scavengers who search for saleable materials. These activities usually scatter the waste that is ready for collection, and this makes the job of the collection crew even more difficult, as they have to shovel the loose and scattered waste from the ground into the collection vehicle. This system leads to unacceptably low collection efficiency and too much waste is left on the streets of Phnom Penh.

3.1.3. Disposal

Collected waste is transported to the only existing dumpsite at Stung Mean Chey (SMC) for disposal. The site has been generally called Stung Mean Chey dumpsite. The disposal method is open dumping. There is no reclamation plan. Waste is merely leveled by bulldozer in a disorderly manner.

The present dumpsite has been used since 1965. It is located in the southeast about 5 km from the city center (central market) and the area is about 6.8 ha, but waste has spread over a larger area possessed by the municipality. It is said that the dumpsite is already full, but because the municipality fund is not adequate to allocate resources for new landfill, collected waste has still been dumped there in spite of warnings about the very high level of pollution. The new landfill project is included in the PIP of the government. However, the project has failed to attract the attention of donors or investment banks.

3.2. Institutional arrangement system

There are some agencies, which are at least partially concerned with SWM in Phnom Penh city. This section reviews first the legal responsibilities of the agencies concerned and then their actual responsibilities in practice.

3.2.1. Responsibilities by law of the concerned agencies

3.2.1.1. Ministry of Environment (MOE). The MOE was established in 1993. The environmental protection and natural resource management law clearly prescribes that the Ministry has responsibility for establishing proper guidelines for SWM and supervising its execution. The MOE is also responsible for monitoring and enforcing compliance with the environmental law and the operation and discharge permits. Therefore, the ministry has the responsibility for SWM at the national level.

3.2.1.2. Ministry of Planning (MOP) and Ministry of Economy and Finance (MOEF). The MOP and the MOEF are involved in approval of sector investment plans, establishment and approval of cost recovery levels and appropriate service tariffs, allocation of government funds for investment projects, and solicitation and organization of grant and loan financing from international financial institutions and donors.

3.2.1.3. Municipality of Phnom Penh (MPP). The subdecree on solid waste management prescribes that the collection, transport, storage, recycling, minimizing

and dumping of waste in provinces and cities is the responsibility of the authorities of the provinces and city. Therefore, the MPP has the responsibility for SWM in the city.

3.2.1.4. Phnom Penh Waste Management (PPWM). MPP established two service organizations in February 2001, namely Cleansing Authority of Phnom Penh (CAP) and the Wastewater Authority of Phnom Penh (WAP). Both were established based on the government sub-decree of April 1999 regarding the government's public administrative financial policies. To reduce the administrative costs and increase operational efficiency, the MPP decided to merge the two organizations into one authority, Phnom Penh Waste Management (PPWM). It is described in a statute of PPWM that PPWM has the responsibility for the solid waste business in the city. According to its statutes, PPWM has very broad mandates; it may itself deliver SWM services, such as collection and disposal, or contract out these services, and monitor the performance of the service providers.

3.2.1.5. Private company. The solid waste service has been provided in Phnom Penh city by a private company. The contracted company has the sole right to collect and dispose of all municipal waste from within the city. The company is also requested to collect user fees from households, enterprises, institutions, etc., to recover its operational costs.

3.2.2. Responsibilities of the concerned agencies in practice

In the present SWM system, PPWM, which has a wide mandate in SWM to control the operation performance of the service provider (private company), is presently only responsible for the operation of its own disposal site, without any permit from the MOE. The MOE, which is responsible for monitoring and enforcing the environmental law, seems unable to perform completely its task, although it admits that the operation at the disposal site by PPWM is neither technically nor environmentally acceptable.

3.3. Existing financial arrangement system

The MPP does not have adequate financial resources to operate its own collection and disposal systems. It has solved this cash flow problem by franchising out the SWM services to the private company, which can recover its cost from people who benefit from the service. In this way, the solid waste service can be offered in the city but the service quality control has not been fully implemented because the MPP has yet to insure financial resources for monitoring the performance of the service provider. It should be noted that the MPP created the PPWM but has not allocated any funds to the body for operation. The revenue of PPWM is mainly from the disposal fees levied on the private company, with a small part from grants from international donors.

4. Evaluating the existing SWM system

In Cambodia, there are some areas in the city where residents are considered squatters. The SWM in these urban poor areas requires an appropriate management strategy that may be different from that for the central urban areas (Kum et al., 2004). This paper is focused only on how to improve the SWM system in central urban areas of the city.

4.1. Evaluating the technical arrangement system

4.1.1. Waste storage

At residential areas, because of an unavailable storage system, waste is placed in plastic bags and left along the curbside for collection. The waste is scattered by scavengers. According to Flintoff (1984), the cost of removing waste which has been scattered in the streets is much higher than the cost of collecting similar waste which has been placed in containers such as domestic waste bins or litter containers. At the commercial and institutional areas, waste is normally placed in large non-standard bins provided by the individual owners. The transfer of waste from the large bins and containers to the collection vehicles is done manually. The process is time-consuming and results in much littering. Although the crew attempts to sweep up the waste, there is a limit to how much time they can spend on this. At the places where the waste needs to be collected daily, or even more frequently, such as food markets, communal collection points, etc., the storage bins are not adequate enough. It is common to see the market waste dumped on the ground in free space near the market and these places become collection points. This practice is technically unacceptable. However, it is still seen continuing in the city. Thus, the lack of a workable storage system at the source of waste generation is considered the most serious shortcoming to the present waste situation in Phnom Penh. An appropriate storage system should be established and introduced. The non-standard individual bins of the owners of the restaurants, hotels and institutions should be replaced with standard bins or containers. This system may need trucks with special loading equipment because the larger standard bins are too heavy to be manually loaded into the trucks. Such a system will increase the investment cost, but may result in operating cost savings as the loading will be much faster and the productivity of the collection equipment and crew will be higher. When the appropriate storage system is introduced, the private

company will need to establish a proper plan for collection according to the volume of bins provided to avoid the spillage of generated waste.

It is also worth noting that while a standard container system is probably significant to improve collection productivity, it also faces some problems to be solved. The fact is that containers for waste storage may be stolen and used for other purposes. This happens in many developing countries (Cointreau, 1982; Flintoff, 1984; Ogawa, 1989). But, the problem may be solved by labeling each container, or by fixing the containers to a post or metal chain, or in any way making the bins easily recognizable as bins for waste storage and not for other purposes. However, without an appropriate storage system, most people still continue their practice of putting waste in plastic bags along the curbside for collection. This will require fixed collection schedules and much more reliable collection timing than today. Therefore, an appropriate storage system can be considered also as a way to facilitate the collection operation and it should be introduced into the waste management system in the city.

4.1.2. Waste collection and transportation

Collection efficiency in the central urban areas under the present operating system is low. The low efficiency may be because the collection crew spends so much time in collecting the loose waste that is normally dumped on the ground or scattered around the scarce rubbish bins, which is due to the lack of an appropriate standard of storage system, and/or because the number of the vehicles used is not adequate for the operation (Mori, 2000).

According to Bhat (1996), the collection and transfer cost normally takes from 70% to 80% of the total budget for solid waste management. Because solid waste collection is a very costly service and the most expensive phase of waste management, the poor technical arrangement of the waste collection system practiced by the private company in the city may lead to a high operation cost. As a result, the private company may try to increase the cost of collection fees to recover the operation cost, but this may not be possible because the per capita income of the general public is still low. As a result, the private company may perform its service according to the resources available that it can manage from the collection fee, and the collection service will deteriorate. As mentioned earlier, PPWM cannot do its job well, monitoring the service performance offered by a private contractor that is in a monopoly situation, especially when the contract with the private contractor lacks specifications with respect to service level and standards. As a result, there may be a potential threat from the uncollected waste to urban environmental degradation in general. Therefore, the existing collection system should be replaced with a more efficient, but not more expensive system.

Waste scattered from the collection trucks during transportation may be due to the lack of appropriate cover during the trip. It is a normal requirement that the waste should be covered during transport and this is imperative for motor vehicles traveling at 30 km/h or more (Flintoff, 1984). So, the waste collection crew should carefully cover the waste during the transportation. However, the use of compactor trucks for the transportation of waste as a means of enclosing waste during transport, may not be an appropriate solution. A compaction vehicle is specifically designed for the waste volume reduction. In developed countries, the compacted density of municipal solid waste is about 400–500 kg/m^3 from the initial density of 100–150 kg/m^3 (Flintoff, 1984). The non-compacted (initial) waste density in Phnom Penh city is already in the range of 350–400 kg/m^3 (Table 4). This means that the benefit from compaction would not be significant. Moreover, the use of compactor trucks requires high investment and operating cost and also complex additional maintenance. It should also be noted that compaction of mixed and wet waste that is typical in Phnom Penh city tends to force out the moisture and discharge it as a leachate. As a result, using compactor trucks to solve the scattering of waste during transport is not an appropriate alternative.

4.1.3. Waste disposal

Because the MPP has very limited financial resources, uncontrolled dumping may be the only option available at the moment because it is the cheapest type of land disposal. However, it should be noted that such a practice puts the public and the environment at risk from underground and surface water contamination, toxic smoke and waste blown by the wind, vectors, etc. Such a practice should not continue because it is not environmentally acceptable, and it makes the useful life of a disposal site even shorter. The economic reason for taking care that disposal sites have the longest possible life is that, once these sites are filled new ones usually can be found only at a greater distance and this increases transportation cost considerably, which accounts for the major share of overall cost (Komorowsky, 1983). Still, an effective strategy to make the disposal sites have the longest possible life should focus not only on technical operation at the site, but also on waste diversion that will include source reduction, recycling, and waste transformation through composting. Unfortunately, the waste management system in Phnom Penh city has not included any strategy to divert the waste from entering the dumpsite. Though recycling and composting can theoretically divert a certain amount of waste from disposal, their chance of success in practice in Phnom Penh city is limited (Kum et al., 2004). So, "prevention is better than cure" may be a good idea in managing the solid waste system. This means that with the same objective to

minimize the pollution from solid waste, it is better trying to *prevent* the waste at source of generation through source reduction than just trying to *cure* the generated waste through waste treatment/transformation.

Regarding waste disposal by incineration, the municipality should not enter such a scheme because the system may not be economical or sustainable to be applied in the city of Phnom Penh. Without conducting a detailed economic analysis, the waste characteristics in the city already shows an important indicator. According to Rhyner and colleagues (1995) "fuel with a high heat value (greater than 5000 kJ/kg), low moisture content (less than 50%) and low ash content (less than 60%) can be burned without additional fuel". The waste characteristics in the city (Table 4), do not meet the specification. Therefore, the MPP should leave this issue to promoters from the private sector to finance, build and operate such plants. If the promoters feel that this system is truly financially sustainable in the country, they should be willing to develop and operate it without subsidies from the Municipality.

4.2. Evaluating the institutional arrangement system

It generally proves to be politically and practically difficult for a regulatory governmental unit to enforce compliance with regulations, which were violated or ignored by other governmental units. Therefore, the MOE would find it difficult to monitor the disposal site management operated by PPWM and it may be difficult/impossible for the MOE to collect fines from PPWM for any violation of regulations.

Also, during the data collection, it was found that the information seems really "*secret*". Data that is available at the PPWM department is not available at the MOE and vice versa. There seems to be a lack of coordination among the relevant waste agencies. This is seen to be one of the major shortfalls in the institutional arrangement system and may lead to the duplication of efforts on the same project because a local waste authority is not aware of what others are doing. This is always a waste of resources and time.

Another important factor that should not be overlooked is the lack of human resources in the PPWM department. According to Mori (2000), there are just a few persons in the PPWM department who empirically have an understanding of waste management, and only one person who learned the SWM systematically through the training course hold by JICA in Japan. This shows that human resources in this field are still very low and more training is needed. Though such a problem is commonly found in developing countries due to the low priority given to the waste management sector (Ogawa, 1989), it proves to be a major obstacle to effective waste management. This is based on the fact that developing countries usually seek external aid packages from donor countries, usually developed countries, to build a Master Plan for almost any project. Only urban SWM case is cited as an example herein. In this case, the consultants are often from the donor countries. The consultants from developed countries sometimes have no background in urban SWM in developing countries and try to promote highly mechanized systems that are successfully applied in developed countries but not in developing countries. This happened in Tanzania. In the Dares Salaam Master Plan (1979), an action program written by the professional who had developed the Master Plan wrote: "Compactor lorries should continue to replace side loaders for the roadside collection. The refuse can be compacted to a density of approximately at 350 kg/m^3 thereby giving a lorry capacity in order of 5,250 kg." (Yhdego, 1988). According to Yhdego (1988), many solid wastes in Tanzania already have an initial density similar to that of compacted waste. This means that the justification in the Master Plan for compaction equipment on the refuse collection vehicle is not reasonable. Such a mistake should not be repeated in Cambodia. But, how can this be avoided? Without adequate competent local human resources in the SWM field, such a problem may be easily repeated in Cambodia. Therefore, human resource development with technical expertise in this field is really needed and should be timely.

4.3. Evaluating the financial arrangement system

Taking into account, the fact that the MPP's financial situation is difficult and the fact that the government's development objectives have prioritized public spending on agricultural and rural development (MOP, 2002b), private sector participation in SWM may be the only realistic alternative to solve the cash flow problem of the MPP. This is based on the fact that, though the SWM project may be considered to have a high priority by the MPP, the project does not fall within the prioritized national development projects and, therefore, it may be difficult to be approved by the central governmental units.

Borrowing money from a development bank may be another alternative, but that, too, is not promising. This is because the preparation of projects to be supported by international development banks will need to follow rather strict and lengthy procedures and requirements (Cointreau, 1982). The tough conditions imposed by the banks may not suit the municipality.

Although private sector participation is viewed as one way to secure investment finance from private companies for solid waste equipment and facilities in return for contracts to provide service, it is very unlikely that the most cost-effective services have been achieved since the contracting was done without competition at the bidding stage. It is believed that better service may be

obtained for the same cost if a competitive bidding process is combined with a service agreement that includes a clear and monitorable service standard. Moreover, the MPP created PPWM but has not allocated any budget to the body to perform its task. The budget for sustaining the body is partly from revenue from disposal fees for the disposal site maintenance allocated by the private corporation, and some small amount from international donors. It should be noted that donor agencies have their own upper limits to financial support and not all donors are interested in the SWM sector. As a result, there is a limited amount of funds to be allocated to the sector (Ogawa, 1989). So, the financial arrangement of the PPWM should be reviewed to better reflect the long-term objective, taking into account PPWM's statute according to which it has a broad mandate and very important role to play in the SWM sector.

The MPP should allocate enough funds for PPWM to perform its tasks, but the promotion of PPWM to be a self-financed institution should be based on a detailed study. This is because the solid waste service may be different from water supply or electricity supply services. Former governmental units in the public sectors, such as Phnom Penh Water Supply Authority (PPWSA) and Electricité du Cambodge (EDC) have developed themselves to be autonomous and self-financed authorities with a remarkable success. However, the strategy might not be promising for the case of solid waste service because the general public still has a low environmental awareness and a low income per capita. Therefore, the willingness to pay the collection service fees may not be as high as to pay the water and electricity fees.

4.4. Public participation

Although the technical, institutional, and financial arrangements of the existing SWM system need improving, public participation is also important and should not be overlooked.

In Phnom Penh city, it is common to see people throw litter from cars or motor vehicles into streets while traveling and to see people in the parks leave litter on the park benches even though rubbish bins are situated within walking distance. This is really a lack of responsibility of being a citizen and has a negative impact on the cleanliness of the city in general. In more serious cases, in some areas in the city, people deliberately dump their waste into open manholes or drains thinking that it will be carried away with the rainwater, not understanding the clogging and pollution problem this causes. Sometimes, they burn their excess waste in the streets, creating health risks and environmental problems. In other areas, especially medium or higher income areas, the situation is better. People do not burn their household waste, but leave their plastic bags of waste along the streets where the waste becomes scattered by scavengers. The private company always complains about this. According to the operation manager of the private company about why there is still so much waste along the streets in the city, the answer is that the general public does not understand and respect the waste collection schedule of the company and that there is a lack of co-operation from the public to make the city clean. But, the question is whether people are well-informed about the waste collection schedule. Is the waste collection schedule fixed? Are there enough storage bins so that people can dump their waste? If not, where can the public put their garbage – on their premises and wait for the irregularly scheduled waste collection? Does the private company really understand this?

However, activities of urbanites such as throwing the waste into the streets, dumping it into open drains or burning it in the streets are the responsibility of the general public. This also shows that the general public has not been motivated to participate actively in waste management issues. So, there is a need for a greater public participation in lobbying for better SWM in Phnom Penh city. The increased public participation may be carried out by means of all sources of media, such as television and radio networks, as well as newspapers, to increase the public awareness. However, for the long-term objective, such a strategy may not be enough. The environmental issues, including the solid waste issue, should be included in the school curriculum to build human resources for future generations.

5. Conclusions and recommendations

It can be inferred that solid waste management has interwoven and interdependent issues. The issues should be addressed from a system perspective by taking into account the technological, financial, institutional, legal, and socio-cultural factors to determine appropriate policies for the local surroundings. It can also be inferred that although private sector participation in solid waste management is seen as a way to solve the financial constraints and offer the solid waste service to meet the demand of the public in Phnom Penh city, the lack of competent professional staff and adequately designated authority of the local waste authorities are major obstacles to develop, negotiate, manage, monitor, and enforce a sound contract instrument.

The recommendations below are based on the discussions as presented in previous sections:

- Appropriate storage systems at the sources of waste generation should be introduced into the waste management system in the city.
- The capacity of the local waste authorities should be improved and enforced by establishing a human resource development program so that they can have a better control of the SWM system in the city.

- Information sharing among relevant waste agencies in SWM should be much wider to avoid duplication of efforts for SWM projects in the city.
- The role and responsibility of PPWM should be reviewed and clarified as to whether PPWM is a supervising agency or an executing agency in SWM in the city.
- The MPP should allocate an adequate budget for PPWM so that PPWM can better perform its task, controlling the performance of the provided service in the city.
- Regulations regarding littering and improper disposal of solid waste should be formulated, and penalties should be imposed on violators.
- Public awareness about the environment should be increased through environmental education and training program so that the public participation in SWM will be increased.
- A source reduction program should be initiated and strongly promoted because it is a way to address waste prevention and the diversion of materials from the waste stream, and this could contribute to a sustainable SWM objective.

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Book reviews

Frank Woodard, P.E., 2004. Industrial Waste Treatment Handbook, Ph.D. Butterworth–Heinemann (ISBN 0-7506-7317-6)

The author states "that his intention in writing this book was to provide a primary reference for environmental managers working in industry, environmental engineering consultants, graduate students in environmental engineering, and government agency employees concerned with wastes from industry".

This book "explains the fundamental mechanisms by which pollutants become dissolved or suspended in water or air, and explains how different treatment processes work, how they can be optimized and how one would go about efficiently selecting candidate treatment processes". Examples are presented to illustrate both the approach used in solving various environmental quality problems and step-by-step design of facilities to implement the solutions.

Chapters include: *Management of Industrial wastes:* Solids, Liquids and Gases; Fundamentals; Laws and Regulations; Wastes from Industries; Industrial Storm Water Management; Waste Characterization: the waste characterization study, wastes audit, and environmental audit; Pollution Prevention; Methods of Treating Wastewaters from Industry; Treatment and Disposal of Solid Wastes from Industry; and Methods for Treating Air Discharges from Industry.

The first chapter includes an example analysis of capital, operating and maintenance costs and annualized costs for four optional pretreatment facilities for comparison with the surcharge that the publicly owned treatment works (POTW) would charge. In addition, management of solid wastes from industry and discharges to the air is briefly reviewed.

Fundamentals of industrial wastewater treatment are briefly reviewed. Laws and regulations concerned with industrial waste treatment are listed and summarized as of 1993, and references are given. Fourteen industries are described as representative of various types of manufacturing processes, with many schematics. Waste minimization, pollution prevention and end-of-pipe treatment methods are discussed. The chapter on waste characterization has an extensive bibliography including USEPA documents.

Chemical, physical and biological methods for treating industrial wastewaters are described in detail. Equalization techniques, pH control, chemical treatment, coagulation, chelation, oxidation and reduction, thermal oxidation and catalytic oxidation are discussed. Design equations for biological treatment systems, and typical calculations are presented: activated sludge systems and their calculation, aerobic and anaerobic systems and their calculation, filtering systems and settling tanks, and mechanisms of ion exchange. Extensive references are provided.

The chapter on treatment and disposal of solid wastes from industry covers landfills and landfill gas, solid waste incineration, and composting. Methods for treating air discharges from industry include: cyclones and precipitators, bag houses, venturi scrubbers and activated carbon adsorption systems. Again, extensive references are provided.

All in all, this volume provides a broad overview of industrial waste treatment. It presents the methodologies available for identifying which waste types are produced from which industrial processes and how they can be treated, neutralized or rendered less harmful and finally disposed safely into the surrounding land, air or waterways. Every environmental professional and plant manager needs to understand the essential principals and processes which are available to assure compliance with government regulations as well as to aid their own consciences that the best methods are applied effectively and economically. Extensive references to the technical literature as well as USEPA documents will assist the reader in searching for the additional information that will be required when he gets into the nitty-gritty of practical engineering and management.

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P.H. Brunner, H. Rechberger, Practical Handbook of Material Flow Analysis, Lewis Publishers, Boca Raton, FL, ISBN 1566706041. 336 p., $120.

The authors wrote this handbook to explain and promote material flow analysis (MFA) as a tool to wisely manage material (and energy) resources from the perspective of environmental protection. The book describes the theory and methods of MFA and its utility and application by way of numerous examples. The examples discussed include "systems" of various types, complexities, and breadth, e.g., industrial resource management, municipal waste management, and nutrient management. The intended audience of the handbook includes engineering students, professional practitioners, and decision-makers.

The book consists of four chapters (each with a separate set of references) and index. Additionally, Chapters 1 through 3 contain sample problems at the end of the chapters.

The introductory chapter defines MFA, which is an accounting of the material inputs, outputs, and inventories (i.e., storage or stock), and describes some of the history and applications of materials (mass) balances in human endeavors. One of the earlier applications was the attempt by an Italian doctor (Santorio Santorio) in the 17th century to measure and close the metabolic mass balance for a human subject.

The main content of the handbook is presented in the second and third chapters. The theory and methods of MFA are presented in detail in Chapter 2. Important terms of MFA and their definitions are presented in this chapter. The procedures for determining the analytical (i.e., system) boundaries and relevant boundary conditions are also described, as well as the methods and rationale for identifying the types and flow patterns that are relevant to the type of resource or environmental analysis under consideration. Material flow and their properties (e.g., heavy metals concentrations) are introduced in Chapter 2 to acquaint the reader with the types of inputs and outputs used in the modeling analysis, and the types of analyses that the evaluator can perform. Three software packages are described by the authors for use in MFA, including detailed discussions of two commercial software products. The discussion includes step-by-step procedures, as well as figures depicting construction and labeling of example system configurations. (No MFA software products are included with the handbook.) A discussion of methods of interpreting MFA results concludes Chapter 2. A number of performance parameters for MFA are described, including material-intensity per service unit, sustainable process index, and Swiss Ecopoints.

Fourteen MFA case studies compose the third chapter. The case studies describe the inputs, outputs, and storage terms for the systems selected for analysis, as well as the methods, results, and conclusions (if any) of the analyses. The case studies cover environmental management, resource conservation, solid waste management, and regional materials management. The complexity of the systems composing the case studies extends from a single facility processing construction waste, to the Bunz Valley region of Switzerland (66 km^2 and 28,000 inhabitants). The Bunz Valley analysis examines the flow of the chemical element lead among subsystems of the region (e.g., households, forest soils, rivers, industry, and landfills).

The Bunz Valley regional analysis demonstrates the multi-disciplinary nature of MFA, i.e., knowledge, data, information, and other support are required from a number of professional fields. The analysis also illustrates that the practitioner must be ready to identify the key variables and conditions governing the analysis in order to simplify the computations and/or circumvent a lack of reliable data.

The wide disparity, complexity, and number of the case studies provide the authors with the opportunity to present and demonstrate their point that humans are improperly managing and conserving material resources, with some profound environmental implications for future generations. MFA provides them with the tool to convey their message in a quantitative fashion.

Among their concluding remarks, the authors state that material flow analysis is a developing and dynamic science. In this regard, in the fourth and concluding chapter, the authors note that the results of MFA are only as accurate as the input data, some of which are difficult, expensive, or both to obtain. They also lament that many industries and institutions do not recognize the utility of MFA in optimizing resource use and management of materials. MFA can be further developed in the future through application of standardized definitions, methods, and procedures and through legislation that would base management of the environment, resources, and wastes on MFA.

For students and professionals interested in MFA and its applications, this handbook is a good introduction. In addition, the compilation of input data and results among the fourteen case studies provides the reader with a comparative context regarding the magnitudes of materials use and associated potential environmental consequences that he or she may not be able to otherwise find in one document. While environmental impact analysis is beyond the capabilities and scope of MFA and this handbook, the potential environmental consequences of the chemical loadings and accumulations presented in the case studies will not be lost on the reader.

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Waste Management Instructions for Authors

Waste Management is an international journal devoted to the presentation and discussion of information on the generation, prevention, characterization, monitoring, treatment, handling, reuse and ultimate residual disposition of solid wastes, both in industrialized and in economically developing countries. The journal addresses various types of solid wastes including municipal (e.g., residential, institutional, commercial, light industrial), agricultural, and special (e.g., C&D, health care, household hazardous wastes, sewage sludge).

Waste Management is designed for scientists, engineers, and managers, regardless of their discipline, who are involved in scientific, technical and other issues related to solid waste management. Emphasis is placed on integrated approaches. These approaches require the blending of technical and non-technical factors. Although the dissemination and application of innovative technical information is extremely important, the implementation of sustainable waste management practices also requires a thorough understanding of the pertinent legal, social, economic, and regulatory issues involved.

The journal strives to present a mix of subject matter that will best serve to help the reader understand the breadth of issues related to solid waste management. The following are some of the major areas in which papers are solicited (further information is provided on the journal's website):

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Treatment (mechanical, biological, chemical, thermal, other)
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World Health Organization, Regional Office for Europe, 1991. Urban Solid Waste Management. Edited by Institute for the Promotion of International Health Actions (IRIS), Copenhagen, Denmark.

Diaz, L.F., G.M. Savage, and C.G. Golueke, 1992. Resource Recovery from Municipal Solid Wastes: Vol. I, Primary Processing. CRC Publishers, Inc., Boca Raton, Florida, USA.

Lasaridi, K.E. and E.I. Stentiford, 1998. A Simple Respirometric Technique for Assessing Compost Stability. Water Resources 32, 3717–3723.

Pohland, F.G., 1986. Sanitary Landfill Stabilization with Leachate Recycle and Residual Treatment. EPA-600/2-75- 043. U.S. Environmental Protection Agency, Municipal Environmental Research Laboratory, Cincinnati, Ohio.

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CONTENTS

L.F. Diaz	1	Editorial
M. Jang, S. M. Hong and J. K. Park	5	Characterization and recovery of mercury from spent fluorescent lamps
P. L. González-Torre and B. Adenso-Díaz	15	Influence of distance on the motivation and frequency of household recycling
P. Jain, H. Kim and T. G. Townsend	25	Heavy metal content in soil reclaimed from a municipal solid waste landfill
M. C. Shinzato and R. Hypolito	37	Solid waste from aluminum recycling process: characterization and reuse of its economically valuable constituents
B. C. O'Kelly	47	Mechanical properties of dewatered sewage sludge
E. Hsu and C.-M. Kuo	53	Recycling rates of waste home appliances in Taiwan
H. M. Veit, T. R. Diehl, A. P. Salami, J. S. Rodrigues, A. M. Bernardes and J. A. S. Tenório	67	Utilization of magnetic and electrostatic separation in the recycling of printed circuit boards scrap
T. Van Gerven, D. Geysen, L. Stoffels, M. Jaspers, G. Wauters and C. Vandecasteele	75	Management of incinerator residues in Flanders (Belgium) and in neighbouring countries. A comparison
V. Bostan, L. H. McCarthy and S. N. Liss	89	Assessing the impact of land-applied biosolids from a thermo-mechanical (TMP) pulp mill to a suite of terrestrial and aquatic bioassay organisms under laboratory conditions
V. Kum, A. Sharp and N. Harnpornchai	101	Improving the solid waste management in Phnom Penh city: a strategic approach
	111	Book reviews

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